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PHYSICS IN INDUSTRY

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PHYSICS IN INDUSTRY

MAGNETISM

*Lectures delivered before the Manchester and District
Branch of the Institute of Physics on
1st and 2nd July 1937*

FOREWORD

THE Conference on "Magnetism" of which an account is given in this book was held in the University of Manchester on 1st and 2nd July 1937, under the auspices of the Manchester and District Branch of the Institute of Physics. Similar conferences have been arranged in the summer term for many years, with the object of bringing together scientific workers in industry and at the University. The common practice in the past has been to select some branch of physics in which progress has been rapid, and to arrange a series of talks, given by guests whom we invited from the universities, which summarized recent advances in knowledge.

It has been our experience that such talks are widely appreciated. It is very difficult to keep oneself informed about any rapidly advancing branch of science on account of the vast number of the papers published in many journals. An expert summary enables one to separate the wheat from the chaff, to judge what is really important and what is trivial, to see the broad trend of modern thought without being confused by masses of technical detail. The talks at previous conferences were planned with this end in view, and the speakers whom we invited were asked to frame their summaries on broad general lines.

This year a different scheme was tried, for both the industrial and university sides were represented. During half the conference we university representatives sat at the feet of the industrial scientists. They were our teachers, and gave us surveys of the knowledge they had accumulated and the problems with which they were faced. It should be stressed that the object of the conference remained what it has always been, to create close links between industry and the universities. We did not arrange a conference at which the industrialists talked to each other, for that would be merely duplicating what is done so well already by their own technical organizations. We asked them to talk to the university workers, to bring themselves down for the time being to our level of understanding of technical matters and to give us a broad general survey which we could follow.

"Magnetism" was a particularly suitable subject for such an experiment. The magnetic properties of materials have been improved in recent years in a most extraordinary way. The theory of

magnetism has also made great strides, yet theory cannot fairly claim any of the credit for the great improvements in magnetic properties of the materials used in industry. These have been made by empirical methods, with practically no guidance from theory at all.

It is probably correct to say that technical advances will always be made by empirical methods, but there is all the difference in the world between pure empiricism, and experiments guided by some theory, however vague and inadequate, in directions which are likely to be profitable. The theory of magnetism has now reached a stage where it ought to be a guide.

One cannot guarantee that the pure scientist, when made acquainted with the problems with which industry is faced, will immediately have a "brain wave" which will enable him to suggest a solution. One can say, however, that unless he has some knowledge of these problems the chances of his being of assistance are very remote. By helping each other to grasp the main essential problems we create the conditions in which inspiration is most likely to come. The results of such a conference may not be immediately apparent, but it creates in people's minds a series of latent images which some chance discovery or line of thought may develop, perhaps after a lapse of a considerable time. "'Tis not in mortals to command success", but we can create conditions which make the prospect of success a hopeful one.

The titles of the papers will indicate the lines taken by the conference. Mr Richer dealt with electrical sheet steel, Dr Dannatt with the limitations imposed on engineering design by the properties of available materials, and Dr Oliver with permanent magnets. On the other side Prof. Mott dealt with the electron theory of magnetism, Dr Stoner with the theory of ferromagnetics, and Dr Bradley (in his paper with Dr Taylor) dealt with the relation between atomic arrangement and magnetic properties.

The publication of the six principal papers was decided upon in response to a general demand for such a record by those attending the conference, and we hope that many others will find these summaries useful. Dr C. Sykes, Honorary Secretary of the Manchester and District Branch of the Institute, has very ably edited the series.

W. L. BRAGG

THE NATIONAL PHYSICAL LABORATORY
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CONTENTS

Lecture I

MAGNETISM AND THE ELECTRON THEORY OF METALS

BY

N. F. MOTT, M.A., F.R.S.

University of Bristol

Lecture I

To the theoretical physicist magnetism counts as a rather difficult subject. The reason for this is, that magnetic forces are so small. One would hardly think so while watching an electromagnet lift scrap iron, but nevertheless it is true from the atomic point of view. Within an atom the electric forces between any two of the electrons of which it is made up are of the order e^2/r^2, where e is the charge on each electron and r is the distance between them; the magnetic forces, on the other hand, are given by $\dfrac{e^2}{r^2}\left(\dfrac{v}{c}\right)^2$, where v is the mean velocity of the electrons and c is the velocity of light. Within an atom, fortunately for those of us who have to understand atomic dynamics, the velocity of the electrons is small compared with that of light, v/c being of the order 0·01. It follows that the magnetic forces are smaller than the electric forces by about 10,000. The forces between atoms are in the main electric in origin, and it is therefore possible to account for the chemical bond, or for cohesion in metals, or for electrical conduction

Fig. 1. (a) Intensity of magnetization of a paramagnetic material, showing saturation at high fields. (b) Intensity for a ferromagnetic material.

without taking into account the magnetic forces at all. Magnetism appears as a small refinement in the theory.

All modern theories of ferromagnetism start from the assumption that the magnetic material contains "elementary magnets"; these are atoms or electrons or something of the kind, the essential point being that each elementary magnet is assumed to have a permanent magnetic moment μ. A gas containing N molecules per unit volume each with a permanent moment μ has susceptibility χ given by Curie's law

$$\chi = N\mu^2/3kT, \tag{1}$$

where T is the temperature and k is Boltzmann's constant. The intensity of magnetization I is proportional to the field H until saturation is approached, when it tends to a saturation value I_0 as shown in Fig. 1. The fields required to produce saturation in paramagnetic materials are very high unless the temperature is low; the

phenomenon has been investigated in Leiden[1] with gadolinium sulphate at $1 \cdot 3°$ K.

The first quantitative theory for ferromagnetic materials was that of Weiss[2]. Weiss made the assumption that in a ferromagnetic body such as iron, there exists a force of non-magnetic origin which over quite a large region constrains all the elementary magnets to point with their axes in the same direction, so that at low temperatures the region is saturated, even in the absence of any external field. An apparently unmagnetized material such as soft iron is made up, according to Weiss, of such magnetized regions or "domains", but with their directions of magnetization pointing in all directions at random. The magnetic field required to twist these directions round so that they are all parallel is very much less than for paramagnetics, a few Gauss for good single crystals (cf. Fig. 1, curve (b)).
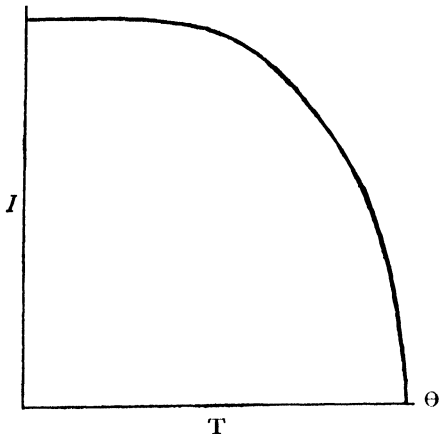
The (non-magnetic) force which constrains all the magnets to point in the same direction has been called by Weiss the intra-molecular field. At low temperatures this force ensures that in any domain the material is saturated. But as the temperature is raised a few of the magnets will be shaken out of their proper alignment, and the intensity I drops below the value I_0 at saturation. But the more magnets are shaken out of their proper alignment the easier is it for the rest to follow, and so, as the temperature is raised still further, the magnetism decreases with a rush and eventually disappears at a well-marked temperature, known as the Curie temperature ($770°$ c. in iron), as shown in Fig. 2. For the mathematical development of these ideas and for their comparison with experiment, the reader is referred, for instance, to Stoner's *Magnetism and Matter* (p. 350).

Fig. 2. Intensity of magnetization of a ferromagnetic material as a function of temperature. The point marked Θ is the Curie temperature.

At the present time we accept, essentially, the Weiss theory as giving the best description of the experimental facts. The advances that have been made since the introduction of the new quantum theory are concerned with (a) the nature of the elementary magnets; (b) the nature of the Weiss intramolecular field.

The elementary magnets are now known to be *electrons*. Evidence mainly from atomic spectra and from the Zeeman effect has shown that the electron is not to be regarded as a point charge but as a more complicated body having a "spin" or angular momentum $h/4\pi$, and a magnetic moment

$$\frac{eh}{4\pi mc} = 9\cdot174 \times 10^{-21} \text{ erg/gauss.}$$

This elementary magnetic moment is known as the Bohr magneton. Moreover, the brilliant work of Dirac[3] has shown that the electron *has* to have a spin and magnetic moment of this amount if it is to obey at the same time Einstein's principle of relativity and the laws of quantum mechanics formulated by himself. We shall not discuss here the evidence, drawn from the gyromagnetic effect, that it is the electron's own magnetic moment which builds up the magnetism of iron, cobalt and nickel, and not any magnetism due to the orbital motion of the electron in the atom.

As regards the hitherto mysterious intramolecular field, Heisenberg[4] has been able to give an explanation of its origin on the basis of quantum mechanics. While this is of course most satisfactory, it does not seem possible to obtain a physical picture of the origin of this field without going rather deeply into the laws of quantum mechanics. We shall content ourselves with pointing out that the field is of the same nature as that which ensures that the two electrons in the molecule H_2 have their spins in opposite directions, so that the molecule can have no magnetic moment unless raised to an excited state.

Heisenberg's work, however, enables certain statements to be made about the force which keeps the elementary magnets aligned, which are of importance for our further considerations. He shows, for instance, that this force is what is called in atomic physics an "overlap" force; that is to say, the orbit of any one of the magnetic electrons in iron must overlap to a certain extent the orbit of the similar electron in the next atom; otherwise the intramolecular field would vanish and the substance would behave like an ideal paramagnetic, as, for instance, the salts of the rare earths, obeying equation (1).

Heisenberg also shows that the intramolecular field can be either positive or negative; if it is negative each elementary magnet will be constrained to point its axis in the opposite direction to that of its neighbours, and the material will normally have no permanent

magnetic moment, as is the case for non-magnetic materials. An intramolecular field may be considered to exist between the atoms of most solids; but only in iron, cobalt, nickel and a few alloys does it have the correct *sign* for ferromagnetism.

The present author has recently considered what part of the atom the electrons come from which are responsible for the magnetism, and this question will be considered next. The shell- or "onion-like" structure of the atom is now well known. In the copper atom, for instance, the electrons can be divided up into two in the K shell, 8 in the L shell, 18 in the M shell and a single valence electron (the $4s$ electron). The K, L, M shells are all "closed shells"; that is to say, they carry all the electrons for which there is room according to the Pauli principle. As a result they are spherically symmetrical, chemically inert and, an important fact for this discussion, they are diamagnetic; that is to say, they cannot have a permanent magnetic moment.

In metallic copper, the valence electrons ($4s$ electrons) come off, and become the free conduction electrons, which form a sort of electron gas extending throughout the metal. Pauli[5] has shown that such an electron gas is only very weakly paramagnetic; we may say if we like that the intramolecular field between the electrons is large and negative.

Owing to the attraction between the positively charged copper ions (Cu^+) and the negative electron gas, the whole assembly tends to contract, so that the spherical copper ions are packed together, like oranges in a case. This at any rate is the picture of metallic copper recently given by Fuchs[6] and used to calculate the elastic constants. For nickel, cobalt and probably iron the picture will be very similar; these elements come immediately before copper in the periodic table, nickel having one fewer electron and cobalt two. Since these metals must have some free conduction electrons in order to be conductors, it follows from simple arithmetic that the M shell cannot have its full number of eighteen electrons. In metallic nickel or cobalt, then, there will be a certain number of vacant places or "positive holes" in the M shell. These will be in the outermost M level, the $3d$ level, which can hold ten electrons. We believe that these "positive holes" are the elementary magnets in ferromagnetic materials.

Compared with the forces holding the atoms of iron or nickel together, the Weiss field gives rise to a rather small force, since the magnetism is destroyed at about 1000° c., while a very much higher

temperature would be required to vaporize the metal. We can see why this is the case in terms of the model given above. The nearly full M shells of the metal behave like rather hard elastic spheres and so cannot overlap very much. The Heisenberg interaction between the positive holes cannot therefore be very great. The valence electron, on the other hand, can come right off the ions, and give rise to very much stronger forces.

The next question which interests us is the number of "holes" per atom. Since at low temperatures we expect all the elementary magnets to be aligned in the same direction, it is reasonable to identify this number with the saturation intensity of magnetization, expressed in Bohr magnetons per atom. This is given in the table below. We give also the number of electrons which have to be distributed between the M shell and the conduction electrons, and since the M shell can take eighteen, a simple sum gives the number of conduction electrons.

	Ni	Co	Fe
Saturation intensity, in magnetons per atom, observed	0·6	1·7	2·2
Number of electrons outside L shell	18	17	16
Number of conduction electrons	0·6	0·7	0·2

It will be seen that in no case is the number of elementary magnets an integer. We must thus think of metallic nickel, for instance, as being a sort of mixture, some of the ions being in the state $(3d)^{10}$ with no positive hole, and some in the state $(3d)^9$ with one. But for most purposes the problem can be treated more fruitfully by using the "collective electron" model, which we shall now describe.

In this model we suppose the electrons to belong not so much to the individual atoms as to the crystal as a whole. This is quite natural for the conduction electrons, but would be very forced for the inner shells; for the M electrons it appears to be legitimate to do so in a qualitative discussion.

We imagine then that in a crystal there are a series of stationary states which are possible for an electron, having energy values lying very close together so that we get practically a continuum. Suppose that an atom has a stationary state which can normally accommodate two electrons; then when N of these atoms are brought together to form a metal, this state will split into $2N$ stationary states. The energies of the states will form a band of levels, which becomes wider

as the atoms are brought nearer together, as is shown in Fig. 3. In hexagonal structures and complicated cubic structures such as that of γ-brass this band of levels is found to split up in a complicated way depending on the crystal structure. These bands of levels are then called Brillouin zones, and it is in terms of them that H. Jones[7] has been able to give an explanation of the phase diagrams of alloy systems which obey the Hume-Rothery rule.* Nothing of this kind happens, however, for the face- and body-centred cubic structures.

Now let us see what will happen in such metals as copper and nickel, where we have to consider both the atomic 3d and 4s levels. In the atom these levels are separated by a few electron-volts only. When the
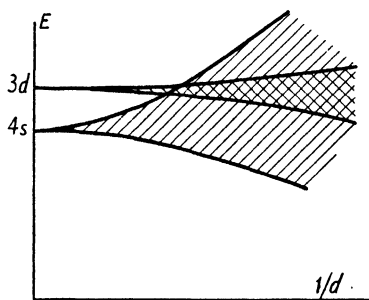

Fig. 3. Energy bands in a metal, as a function of inter-atomic distance *d*.
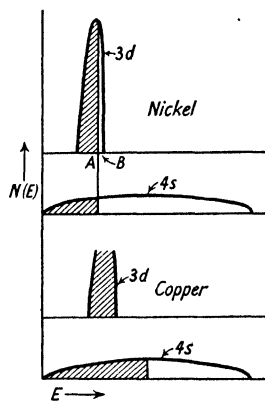
Fig. 4. Density of states in metallic copper and nickel.

atoms are brought together to form the metal both levels will split up into a band of levels, but because the M shells overlap one another only to a small extent, the 3d level will split only into a narrow band, as shown in Fig. 3. In nickel the *s* band will have breadth of about 10 e.v., the *d* band 1 e.v.; but the evidence for this cannot be discussed here. For this reason, and because the number of levels in the *d* band is 10 N as against 2 N in the *s* band, the density of states will be much greater in the *d* than in the *s* band. Fig. 4 shows the density of states $N(E)$ in the two bands, $N(E)\, dE$ being defined as the number of states with energies between E and $E + dE$.

Now according to the exclusion principle of Pauli only one electron can occupy any stationary state, so that at not too high a temperature all the lower states will be occupied and all the higher ones empty. This also is shown in Fig. 4. In copper all the *d* states and half the *s*

* Cf. also ref. (8) for a simplified discussion of this work.

states will be occupied. In nickel, on the other hand, the highest *d* states will be empty.

The model illustrated in Fig. 4 enables one to understand the behaviour of the saturation moments in such alloy systems as Ni-Cu, Ni-Al or Ni-Fe as the concentration is altered. The *substitution* of a copper for a nickel atom will be equivalent to the addition of one electron to the alloy; similarly, an aluminium atom will add three, and an iron atom subtract two. If electrons are added to the alloy they must go into the highest unoccupied states. Thus, for instance, as copper is added to nickel, the boundary between the occupied and the unoccupied levels will gradually rise. When the composition 60 atomic per cent of copper to 40 atomic per cent of nickel has been reached, all the vacant places in the *d* band have been filled, so that the number of elementary magnets has sunk to zero and the magnetism disappeared. Aluminium will fill up the vacant places three times as quickly as copper; iron, on the other hand, *increases* the number of vacant places twice as fast as copper decreases them.

Fig. 5 shows the observed values of the saturation intensities of some alloy systems of nickel, expressed in Bohr magnetons per atom. For concentrations of the second metal which are not too large, the predictions of the theory seem to be quantitatively in agreement with experiment.

The model described here shows that the saturation intensity of an alloy depends in a rather fundamental way on the atoms of which it is built up, and is not likely to be altered appreciably by heat treatment or by the addition of small quantities of any further component. To alter the number of elementary magnets by any large amount one has also to alter the number of conduction electrons, and as the conduction electrons are responsible for the cohesion we see that no small change could bring this about. It seems therefore unlikely that new alloys will be found having a saturation intensity much greater than that of those already known.

The element palladium lies in the same column of the periodic table as nickel. It is highly paramagnetic, having an atomic susceptibility 555 at room temperature. We may assume that this high susceptibility is due to vacant places in the outermost nearly full shell, the N shell in this case, and that the Heisenberg interaction is negative. It is interesting that if palladium is alloyed with copper or silver or gold the paramagnetism drops rapidly and disappears at the same composition as that at which the ferromagnetism disappears in

the nickel-copper alloys, namely 60 atomic per cent of the non-magnetic metal[9]. A similar phenomenon occurs when palladium absorbs hydrogen. We deduce, therefore, that the number of elementary magnets in palladium is just the same as in nickel, namely, 0·6 per atom.

Fig. 5. Saturation moments of alloy systems, in Bohr magnetons per atom.

Up to the present we have dealt with the saturation intensities of magnetic materials. We must now turn to the Curie temperature, namely the temperature at which the ferromagnetism disappears. This depends on the Weiss intramolecular field, or, in terms of more modern ideas, on the Heisenberg interaction between the ions of the metal.

Above the Curie point a ferromagnetic material shows para-

magnetism, and the susceptibility at a temperature T is given in many cases roughly by

$$\chi = \frac{\text{const.}}{T - \Theta},$$ (2)

where Θ is the Curie temperature. Many paramagnetic materials, for instance palladium, have a susceptibility over a wide temperature range of the form

$$\chi = \frac{\text{const.}}{T + \Delta}.$$ (3)

It is convenient to write $-\Delta = \Theta$ and to speak of Θ as the (negative) Curie temperature of material, because Θ, as before, is proportional to the Heisenberg interaction between the ions of the material, which is in this case negative.

What then is the condition that Θ shall be positive, so that a given material is ferromagnetic? Let us imagine that by some method a magnetic material such as nickel could be expanded a good deal, so that the ions no longer touch; then Θ will sink to zero and the material will behave like a paramagnetic obeying Curie's law (1). As the ions are gradually brought together Θ will increase at first; but it has been shown by Bethe[10], by considering the rather complicated mathematical expression which describes the Heisenberg interaction, that Θ after rising to a maximum decreases again and finally becomes negative. The kind of behaviour to be expected is shown in Fig. 6, where Θ is plotted against the distance between the ions, as a multiple of the ionic radius.

The conditions for ferromagnetism are then as follows: firstly the metal must contain ions with incomplete shells; and secondly the ions must touch, but only just. If they are pressed too closely in contact Θ becomes negative and the material becomes paramagnetic. Slater[11], by considering ionic radii calculated by means of wave mechanics, has shown that the latter condition is fulfilled best in iron, nickel, and cobalt.

If a magnetic material lies in Fig. 6 to the right of the maximum, then Θ will be increased and the magnetic energy lowered if the material is compressed. Conversely the presence of spontaneous magnetization will tend to contract the material slightly, so that an anomalous expansion will be observed as the temperature of the material is raised through the Curie temperature. Nickel shows such an effect and must therefore be placed as in Fig. 6. α-iron, on the

other hand, contracts on passing through the Curie point, and so must lie to the left of the maximum as shown. We show α-iron on a different curve, because it has the cubic body-centred structure, while nickel and γ-iron are face-centred. The Heisenberg interaction depends on the number of neighbours as well as on the distance between the atoms.

Fig. 6. Curie temperature Θ as a function of the distance r between the ions, divided by the ionic radius r_0.

Fig. 7. Curie temperature of iron-nickel alloys (observed).

γ-iron, being paramagnetic, must lie on the negative part of the curve. As Slater's work shows, the ionic radius r_0 is greater in iron than in nickel, so that r/r_0 is smaller, although r is much the same. Of interest, then, is the behaviour of the face-centred iron-nickel alloys. Clearly the *mean* value of r/r_0 will decrease uniformly with increasing concentration of iron, and we expect Θ first to rise to a maximum and then decrease. This is in fact observed, as is shown in Fig. 7. Moreover

the position of invar on this curve is of interest. Invar lies where the magnetic transformation temperature Θ is near room temperature, and we see from the slope of the curve that the material will tend to contract as the magnetism disappears. This contraction, balancing the normal thermal expansion, gives to invar its characteristic property.

Alloys with high permeability such as permalloy lie near the maximum of the Θ-concentration curve; it is not our purpose here to discuss the origin of the high permeability; it is probably connected both with a low magnetostriction and with nearly equal ease of magnetization in both crystallographic directions. For alloys lying near the maximum of the curve, Θ and hence the magnetic energy will be only slightly affected by volume changes. Although no theoretical proof has yet been given, it seems quite probable that they would also be unaffected by other strains, in which case the low magnetostriction at any rate would be explained.

In the study of phase diagrams of alloy systems, it may happen that the magnetism disappears at a point on the diagram where there is no corresponding structure change. From what has been said above, it will be apparent that there are two ways in which this can happen. It may be that, while the number of elementary magnets remains finite or even increases, the Curie temperature Θ decreases with changing concentration and drops below the temperature at which the measurements are made, or even becomes negative. The face-centred nickel-iron alloys are an example of this class. On the other hand in alloys such as nickel-copper the number of elementary magnets as well as the Curie temperature tends to zero.

In alloy systems of the former class we shall not expect any discontinuity in the non-magnetic properties of the system at the point where the magnetism vanishes, because, as we have seen, the Heisenberg interaction is quite small compared with the cohesive forces which bind the atoms of the metal together. It is, however, otherwise with systems in which the number of elementary magnets tends to zero. For consider for instance the copper-nickel system, and starting with pure copper suppose that the concentration of nickel is increased. Up to 40 per cent of nickel the number of magnets remains zero, so that the nickel atoms must enter the lattice as closed shells, having the electron configuration $(3d)^{10}$. After this composition each nickel atom contributes just one elementary magnet, and hence one "positive hole"; we must think of the atoms, then, as taking

their places as positive ions with the electron configuration $(3d)^9$, together with one free electron which is added to the conduction electrons. The rate of change of many properties, such as for example the energy, may be expected to be different in the two regions.

An example is provided by the measurements of Lowery[12] and his co-workers on the optical constants of the copper-nickel alloys.

Fig. 8. Absorption coefficient of copper-nickel alloys. *Reproduced by permission from the Philosophical Magazine.*

Fig. 8 shows nk plotted against composition for a constant wavelength.

nk measures the rate of absorption of energy from the light. The wave-length used is in the red, so that the light is strongly reflected by copper, and nk is small. The rise in the absorption as nickel is added is probably due to the ejection of electrons from the closed shell $(3d)^{10}$ of nickel[13]; it will be seen that the absorption coefficient rises only while the nickel atoms are being added in this form.

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Lecture II

THE object of this paper is to give some account of the fundamental physical factors that govern the magnetic performance of commercial electrical sheet material, with special reference to transformer sheet steel; and to indicate briefly the nature of the obstacles that prevent the attainment of magnetic perfection in a reasonably economical commercial product.

The ideal "soft" magnetic medium would be an infinitely large, perfectly pure, and absolutely unstrained crystal of iron, or in other words, an iron crystal perfectly free from lattice distortion of any kind. If we introduce a finite crystal boundary, we inevitably produce some measure of lattice distortion. Even if there is not actual physical distortion of the atomic lattice itself, there will certainly be some measure of magnetic distortion, because, no matter what picture we may form of the elementary magnetic systems associated with the iron atoms, if we introduce a finite crystal boundary, we must necessarily throw some of those elementary magnetic systems out of perfect balance, and so give birth to the hysteresis effect.

With a large, single, finite crystal of pure unstrained iron, it is true that we should still remain very near to magnetic perfection, because the grain-boundary hysteresis effect would not penetrate very far into the crystal structure. But multiply the number of grains in a given volume so as to represent more practical conditions, and even if we imagine only 1000 grains to the cubic millimetre, which is a fair-sized grain for commercial material, it is easily seen that we shall immediately encounter a very serious departure from the ideal, and the appearance of a considerable measure of alternating hysteresis loss.

It would not be difficult in practice, by the proper control of rolling and heat-treating operations, to produce a larger grain size than we do; but, unfortunately, the presence of a certain number of grain boundaries is essential if thin sheet material is going to be physically strong enough to stand up satisfactorily to shearing, stamping, and punching operations, especially when the iron is appreciably hardened and embrittled by the addition of a high percentage of silicon, as in the best qualities of transformer sheet steel.

As an example of the kind of limit that we have to face in practice when we try and work towards magnetic perfection, it is necessary to realize that if we are dealing with a sheet of about $\frac{1}{3}$ mm. (0·014 in.) in thickness, which is the standard in this country for transformer work, and if we have less than an average of about 3 grains of a 4 per cent silicon-iron alloy across the thickness of that sheet, then we are verging towards the limit of practical physical workability. That means that the practical limit of reducing grain-boundary hysteresis loss is beginning to assert itself when we still have left some hundreds of grains in each cubic millimetre, with all their boundaries busily engaged in upsetting our ideal magnetic conditions.

Further, when considering the practicability of improving magnetic sheet material by increasing the grain size, we have also to take into account the effect of eddy currents. Ruder[1] demonstrated quite a long time ago that in thin sheet material, eddy-current losses increase with grain size up to the point at which the grain diameter is equal to the thickness of the sheet. Consequently, if we make our grain size too large, we may easily find that a reduction in the hysteresis loss is counterbalanced by an increase in the eddy-current loss—a very unwelcome phenomenon which has on occasion led quite serious observers to the conclusion that an increase in grain size means an increase in hysteresis loss.

In practice, with a good quality transformer sheet of 0·014 in. thick, giving a total power loss due to hysteresis and eddy currents of 1·15 w. per kg. at B (max) 10,000, and 50 C.P.S., the hysteresis loss due to the grain-size effect will be of the order of 0·25 w. per kg. In a sheet of this quality, the eddy-current loss will also be about 0·25 w. per kg., which leaves us with a total hysteresis component of 0·90 w. per kg. We see, therefore, that the estimated hysteresis loss of 0·25 w. per kg. due to the grain-boundary effect is not far short of 30 per cent of the total hysteresis component. And, at the moment, it would be difficult to contemplate the production of a commercial transformer sheet, 0·014 in. thick, with sufficient physical strength to stand up to even the very best shop practice in stamping and punching operations, without introducing a grain-size hysteresis loss much lower than 0·20 w. per kg. This may seem rather a sad falling away from magnetic grace, but we must console ourselves with the reflection that a transformer sheet with the physical properties of a single crystal would not be of very much practical use.

Other things being equal, hysteresis decreases as the grain size

increases, but it will be obvious that a direct relationship between grain size and hysteresis loss can only be expected to exist when there is a fairly even balance between the principal directions of grain orientation. Suppose, for example, that thin sheet material is produced by processes that involve very heavy cold reduction of the metal, so that the final annealed product reveals marked preferred orientation; then, so far as hysteresis loss due to grain-boundary effect is concerned, it may be possible, in *one preferred direction*, to approximate to the magnetic properties associated with single crystals, even though ordinary micro-examination may indicate a relatively small grain structure. Unfortunately, the excellent magnetic properties which can be secured by such a method of rolling are only recorded in the one preferred direction, and are offset by abnormally inferior properties in other directions.

So far, we have only dealt with what happens when we introduce grain boundaries into a structure of pure metal. Now let us consider what happens when we introduce *impurities*, such as atoms of carbon, oxygen, or sulphur, into the crystal lattice of iron.

If all such impurities could be segregated into nice, well-behaved little lumps, then, by comparison with the grain-boundary effect itself, they would not do a very great amount of harm, because, in a good steel, they would only amount to a small fraction of 1 per cent. In a good 4 per cent silicon steel, for example, the total impurities do not amount to more than about one-tenth of 1 per cent of the iron present. And if we had that percentage fairly evenly distributed throughout the crystal structure in the form of small aggregates, then the magnetic properties of commercial electrical sheet material would be very much better than they are. But with the impurities that we have to contend with in practice, the conditions are very different.

The chief obstacles to commercial magnetic perfection are carbon, oxygen, and sulphur; and these elements all possess the extremely objectionable characteristic of leaving atomic stragglers to create a disturbance inside the iron lattice—that is, in solution—either in true solution, or in the form of colloidal precipitates. And in such a form, even in what might seem fantastically small percentages, these elements, especially carbon and oxygen, can do an almost incredible amount of magnetic damage.

We are indebted to Yensen[2, 3] for practically all of our existing knowledge on the quantitative effect of impurities on the magnetic properties of iron. Fig. 1 shows the effect, according to Yensen, of

small quantities of carbon on the hysteresis loss of single crystals of iron containing varying amounts of silicon. Curve *A* shows the effect of carbon for silicon-free iron. It will be noticed that there is a fairly sharp break in this curve between 0·006 and 0·007 per cent of carbon; and in pure iron, carbon up to this content is regarded as being either

Fig. 1. Hysteresis loss versus carbon. Single crystal conditions.
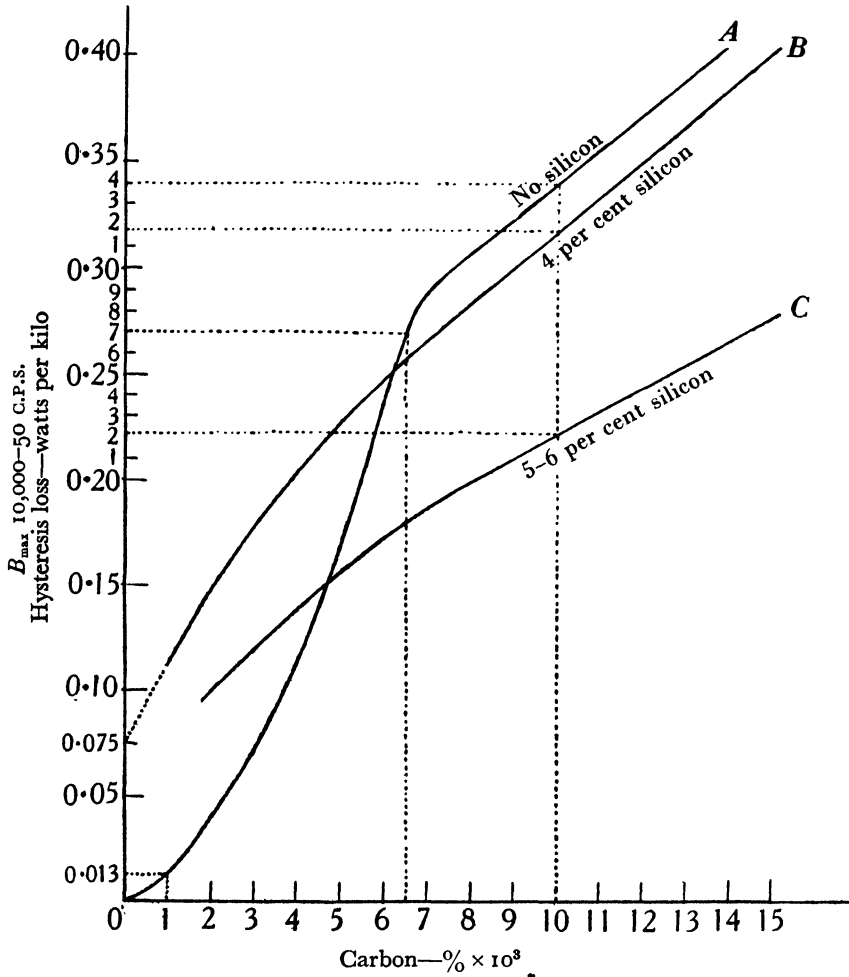
in true solution or in colloidal dispersion. Within this range it is evident that the adverse effect of carbon on the magnetic properties increases very rapidly, the "carbon" hysteresis loss at 0·0065 per cent being 0·27 w. per kg.

Above the 0·007 per cent carbon point, increasing carbon is precipitated in the form of iron carbide, or cementite; and it is clear from

the change in the slope of the curve that the adverse effect of carbon in this form is appreciably less than when it remains in solution. Nevertheless, its effect is still unpleasantly large, the slope of the curve in this range indicating that an increase of 0·01 per cent of carbon increases the hysteresis loss by about 0·16 w. per kg. That may not be very serious if we are dealing with ordinary dynamo iron, but on reference to curve B, which shows the hysteresis loss due to carbon in the presence of 4 per cent of silicon, it will be seen that the slope of the upper portion of this curve is substantially the same as the slope of curve A in the same carbon range—that is, in what we may call the "commercial" carbon range—above 0·01 per cent of carbon.

The only difference between the two curves is that, in the presence of 4 per cent of silicon, the adverse effect of similar quantities of carbon is slightly reduced. But even in the high-silicon alloy, an additional 0·01 per cent of carbon still means an increase in the hysteresis loss of the order of 0·16 w. per kg., and in the best qualities of transformer steel, that means a difference of about 15 per cent in total power losses.

The adverse effect of carbon on maximum permeability is, of course, exactly similar to its effect on hysteresis loss. Fig. 2 is Yensen's curve for the effect of carbon on the maximum permeability of single crystals of pure iron. It will be noticed that the effect of 0·0065 per cent of carbon is to reduce the maximum permeability to 40,000, and that the effect of 0·011 per cent of carbon is to reduce it to 20,000. Yensen estimates that in a single crystal of iron containing only 0·001 per cent of carbon, the maximum permeability should be of the order of 500,000. That is, the difference between 0·001 per cent of carbon and 0·011 per cent means a reduction in maximum permeability from 500,000 to a miserable 20,000. And it will be conceded that there are not many industrial operations where a decrease of 0·01 per cent in the magnitude of an adverse factor makes as much difference as 0·01 per cent of carbon does to the maker of electrical sheet steel.

Fig. 3 is Yensen's curve for the effect of oxygen on the maximum permeability of single crystals of pure iron. It will be seen that the adverse effect of oxygen increases very rapidly up to about 0·02 per cent, and less rapidly between 0·02 and 0·03 per cent. Above 0·04 per cent, additional quantities of oxygen have a relatively negligible effect. It will be noticed, however, that for carbon-free iron, under

single crystal conditions, even 0·02 per cent of oxygen will reduce the
maximum permeability to 110,000, and that 0·04 per cent will drag
it down to 90,000. Not quite so bad as carbon, but quite bad

Fig. 2. μ_{max} versus carbon extrapolated to 0 per cent oxygen
and single crystal.

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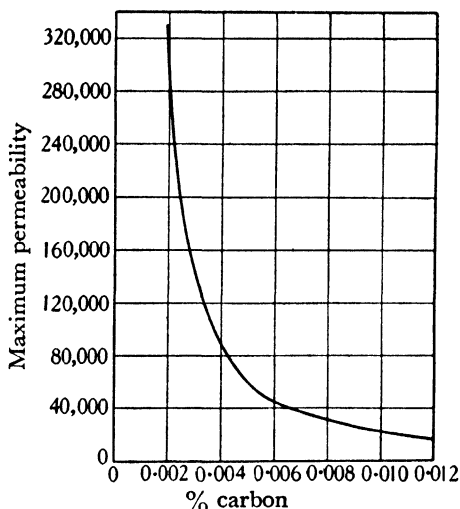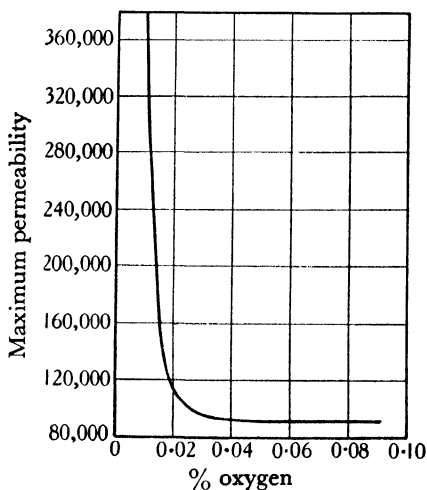

Fig. 3. μ_{max} versus oxygen extrapolated to 0 per cent carbon
and single crystal.

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enough, because it is practically certain that any factor which brings
the maximum permeability down to 90,000 will also bring with it a
hysteresis loss of the order of 0·10 w. per kg.

We are not quite so helpless, however, in dealing with oxygen as we are in trying to deal with small quantities of carbon. The inherent vice of carbon is characteristically human—it reveals itself while it is still young—while it is still in solution. Oxygen betrays exactly similar tendencies. But in the case of oxygen, we are able to do something that we are unfortunately unable to do with carbon. We can keep it from going into solution.

Every schoolboy knows that oxygen has a very great affinity for silicon, and if we add silicon to our molten iron, our oxygen will settle down to a comparatively harmless married state—a typical example of the magnetic advantages of aggregation over dispersion, when it is a question of the effect of impurities. Yensen has established that in the presence of 3 per cent or more of silicon, small amounts of oxygen have no appreciable effect on the low-density magnetic properties of iron-silicon alloys.

But silicon can do other things besides look after oxygen, and we can now consider the general effect of additions of this very useful element to the iron lattice. Briefly, we can describe the benefits of increasing silicon additions to good electrical sheet iron as consisting in a progressive decrease in the hysteresis loss, with its natural accompaniment of a progressive increase in maximum permeability, and in a progressive decrease in the eddy-current losses. For sheet material of similar thickness, the hysteresis loss per cycle in 4 per cent silicon transformer steel will be approximately one-half the hysteresis loss in an ordinary dynamo quality sheet; the maximum permeability will be about twice as great; and the eddy-current loss will be reduced to about one-quarter.

The reduction in the eddy-current loss is, of course, due to the fact that the specific resistance of 4 per cent silicon steel is just about four times as great as the specific resistance of ordinary dynamo iron, being $56\,\mu\Omega$. per cm. cube as against 14.

The reduction in the hysteresis loss is due, not only to the suppression by silicon of the bad effects of oxygen, but also to the larger grain structure that is characteristic of high-silicon steel. It is this natural tendency of high-silicon steel to grow large grains that makes it necessary for us to be so very careful of what we do if we want to arrive at a final sheet product that is neither physically useless nor burdened by excessive eddy-current losses.

Silicon also tends to reduce the hysteresis loss in another way. It will be remembered that the bad effect on low-density magnetic

properties of small quantities of oxygen is suppressed with a silicon content of 3 per cent. Now, we should naturally expect increased additions of silicon to go on doing useful work and still further reduce eddy-current losses by increasing the resistivity of the metal. But additions of silicon beyond the 3 per cent necessary to take care of oxygen have also an interesting effect on the hysteresis loss due to carbon.

Curve *B* (Fig. 1) shows that, with 4 per cent of silicon, the hysteresis loss due to carbon above about 0·007 per cent is reduced by about 6 per cent. For 0·01 per cent of carbon, for example, the hysteresis loss with 4 per cent of silicon is 0·32 w. per kg., as against 0·34 w. per kg. in silicon-free iron. Now, the available evidence does not indicate that an increase in the silicon content tends to keep carbon out of solution as it keeps oxygen out of solution. The reduction in the carbon hysteresis loss in the presence of silicon appears to be due to the fact that as the silicon content increases carbon tends to precipitate in the form of graphite rather than in the form of cementite. And in the form of graphite, carbon is by no means so harmful to magnetic properties as it is in the form of cementite. This is no doubt due to the fact that carbon, as graphite, forms larger aggregates than it does in the form of cementite. This is another example of the magnetic advantages of the "aggregation of impurities".

The tendency for carbon to precipitate as graphite increases with increasing silicon, and curve *C* (Fig. 1) shows the effect of carbon in the presence of a silicon content of 5–6 per cent. The marked decrease in the hysteresis loss which results from the increased silicon content is immediately apparent. For 0·01 per cent of carbon, the loss is about 0·22 w. per kg. as against a loss of 0·32 w. per kg. for 4 per cent of silicon, representing a reduction of 30 per cent. Further, the reduction in the slope of curve *C* as compared with the slope of curve *B* indicates that, in a 5–6 per cent silicon steel, the effect of an additional 0·01 per cent of carbon is only about two-thirds of the effect of a similar addition of carbon to a 4 per cent silicon material.

Unfortunately, hard physical reasons prevent us from employing silicon contents much above 4 per cent. As we approach a silicon content of about 4·5 per cent, there is a most disappointing falling away in the ductility of the iron-silicon alloys. They become difficult to work even under hot-rolling conditions, and they are certainly quite impossible to handle in the cold. This is a very great pity, because, with a 5–6 per cent silicon steel, we could secure an im-

provement in the total power losses in the best qualities of transformer steel of the order of 10 per cent, and such an improvement would be very welcome indeed.

But in spite of the improved magnetic properties that the use of silicon will give us, we must not forget that silicon is an impurity in the iron lattice, and, like all impurities, is inherently harmful to the magnetic properties of iron.

Let us refer once again to Fig. 1. It will be noticed that curve A for silicon-free iron comes down in a quite well-behaved manner to the origin, but that curve B, for iron containing 4 per cent of silicon, does nothing of the kind. And we may reasonably conclude that the intercept on the hysteresis ordinate represents the measure of the harm that 4 per cent of silicon does to the pure iron lattice. The approximate hysteresis loss due to the presence of 4 per cent of silicon is therefore about 0·075 w. per kg., that is, about 10 per cent of the hysteresis component of the total power loss in a really good transformer sheet. We may indirectly secure a net improvement in magnetic properties by the use of silicon, but we obviously have to pay fairly heavily for it.

Further, the progressive addition of silicon involves a progressive decrease in saturation values. For ordinary dynamo iron, the saturation value is about 21,500, but for 4 per cent silicon steel it falls to about 19,500. This is a reduction of approximately 9 per cent, made up of a 4 per cent decrease in the number of iron atoms present, and of a 5 per cent decrease in the physical density of the 4 per cent alloy as compared with unalloyed iron. And since the low-density permeability of 4 per cent silicon transformer steel is appreciably superior to that of ordinary dynamo iron, this reduction in the saturation value of high-silicon steel inevitably involves a crossing-over of the normal magnetization curves in the higher induction range, with the result that the high-density permeability of the 4 per cent alloy is markedly inferior to that of the ordinary dynamo quality, as illustrated in Fig. 4. This inferior permeability at the higher inductions may not worry us very much in practice, because the graduated addition of silicon in electrical sheet steel is expressly designed to produce a differential balancing of electrical and magnetic properties to meet the variation in working induction conditions in different types of electrical machines. In certain types of rotating machines, for example, high-induction permeability is much more important than total power losses; but in transformer work, it is

reduction in total power loss that comes first, although it would undoubtedly be very pleasing if we could combine low power losses with improved permeability in the higher induction range.

The adverse effect of sulphur on magnetic properties is not so great as that of carbon, but it is nevertheless quite appreciable.
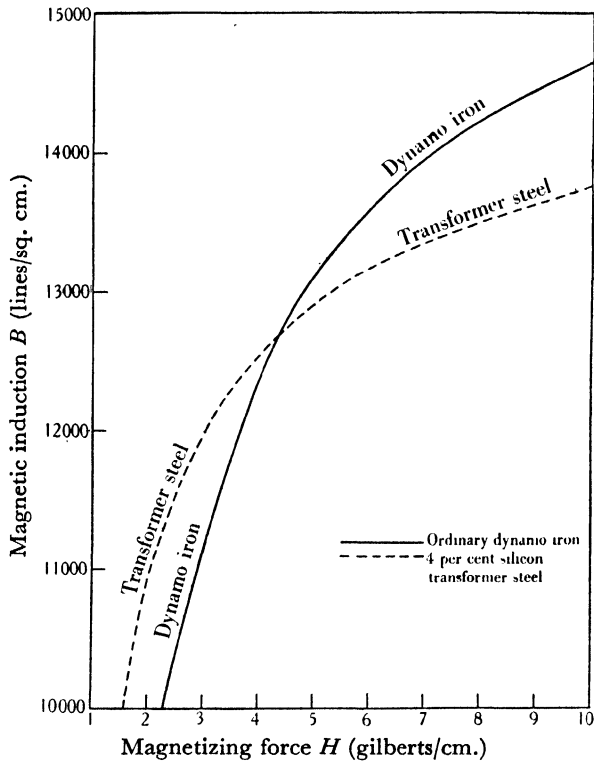

Fig. 4. Normal induction curves.

We have not so much detailed data available on the magnitude of the sulphur effect as we have in the case of carbon and oxygen, because research work similar to that carried out by Yensen has been performed chiefly on material prepared from electrolytic iron practically free from sulphur, phosphorus, and manganese. Yensen has found, however, that the bad effect of sulphur increases in linear fashion with increasing sulphur content, and that it is not quite so great in 4 per cent silicon steel as in iron free from silicon. He estimates that 0·012 per cent of sulphur, for example, is responsible for a hysteresis loss of about 0·14 w. per kg. in silicon-free iron, and of about 0·10 w. per kg. in 4 per cent silicon steel. Now, in the 4 per

cent alloy, under single crystal conditions, a total carbon content of 0·012 per cent is responsible for a hysteresis loss of about 0·35 w. per kg. (Fig. 1). We see, therefore, that for carbon and sulphur contents of 0·012 per cent each, the adverse effect of the sulphur is about 30 per cent of the adverse effect of a similar quantity of carbon; and that if sulphur could be completely eliminated, we might secure another improvement of the order of 10 per cent in the total power losses in transformer sheet steel
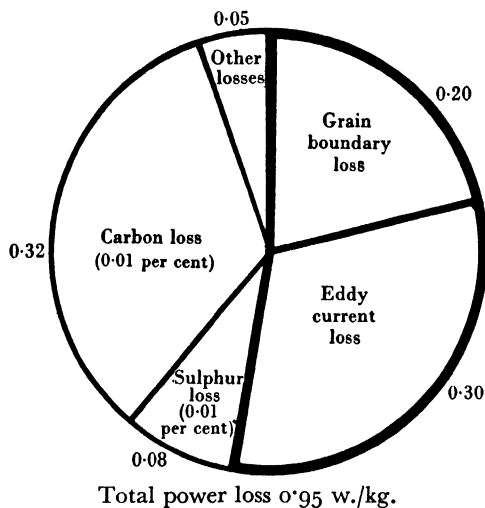

Total power loss 0·95 w./kg.

Fig. 5. Relative importance of losses in 4 per cent silicon steel 0·014 in. thick. (B_{max} 10,000–50 C.P.S.)

So far as other impurities such as phosphorus and manganese are concerned, their combined effect on the hysteresis loss in good quality transformer steel can safely be estimated as not exceeding 0·05 w. per kg.

Now let us see just where we stand. We have seen that it would be physically impracticable under ordinary conditions to reduce our grain-size hysteresis loss to much less than 0·20 w. per kg. It would certainly be economically impracticable at the moment to contemplate either carbon or sulphur contents below 0·01 per cent, which would give us another 0·40 w. per kg. Add another 0·05 w. per kg. for other impurities, and at least 0·30 w. per kg. for eddy-current losses, and we arrive at an optimum total power loss for 4 per cent silicon sheet steel 0·014 in. thick of about 0·95 w. per kg. at B (max) 10,000 and 50 C.P.S. Fig. 5 shows at a glance the relative importance of the various adverse factors that have been discussed. This order of things

fairly represents the results of the best transformer-sheet-making practice to-day, even with expensive refinements in production methods; and there is no maker of transformer sheet steel who would not feel that he was "walking on air" if he could say that the general run of his results would consistently reproduce that figure.

This discussion will have made it quite clear that improvement in practical magnetic performance depends on success in two possible lines of development. In the first place, we can study the possibility of improving the purity of our material. But when we are already dealing with a product in which the total impurities do not amount to more than one-tenth of 1 per cent of the total iron present, the problem before us is obviously not an easy one. In the second place, we can consider whether it is possible to reduce the grain-boundary hysteresis effect without sacrificing essential physical properties, without introducing a very disturbing lack of uniformity in directional magnetic properties, and without cancelling the greater part of our gains by an unwelcome increase in eddy-current losses. And that part of our problem does not even sound as though it were capable of simple solution. We do not admit, however, in spite of all the apparent difficulties, that we have ever reached finality in the magnetic quality of electrical sheet material.

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Lecture III

THE INFLUENCE OF THE PROPERTIES OF AVAILABLE MAGNETIC MATERIALS ON ENGINEERING DESIGN

BY

C. DANNATT, D.Sc., M.I.E.E.

Research Department, Metropolitan-Vickers Electrical Co. Ltd.

Lecture III

I N spite of the progress made in recent years in the physical outlook
on magnetism, and in the developments on the metallurgical
side which have led to new magnetic materials of the greatest value
in certain specialized applications, there has not been any very marked
advance in the magnetic properties of materials used for heavy
electrical plant for many years. Yet it is here, for rotating machines

Table I. Range of properties of commercially available magnetic materials

	Saturation B-H gauss	Electrical resistance microhms per cm. cube	Hysteresis loss ergs c.c. per cycle	Total loss w. per kg. 50 cycles	Permeability	
					μ max	μ_0
Dynamo sheet (0·016 in.)	21,100	14	2900 (B 10,000)	3·0 (B 10,000)	5000	200
1·5 % silicon sheet (0·016 in.)	20,200	30	2400 (B 10,000)	2·1 (B 10,000)	6000	300
4 % silicon sheet (0·014 in.)	19,500	60	1700 (B 10,000)	1·3 (B 10,000)	7000	500
Ni, Fe (mumetal) (0·014 in.)	9,000	40	5 (B 1,000)	0·007 (B 1,000)	6×10^4	12000
Ni, Fe, Co (perminvars)						
Type 1						*1400
Type 2						†100
Dust cores		$2-100 \times 10^5$	0·003 (B 10)			20–50

* This value is constant from $H=0$ to 0·5 gauss.
† This value is constant from $H=0$ to 4 gauss.

and transformers, that by far the largest quantities of magnetic
material are used. Improvements in design, which have been many,
have been mainly due to the better utilization of existing properties.
   The characteristics of magnetic materials of primary interest to
engineers are permeability, losses under alternating magnetization,
mechanical properties, and costs. In the majority of applications
the magnetic material carries alternating fluxes, making laminated
construction essential. Consequently these remarks deal mainly with
materials· in sheet form. Table I lists the principal properties of
typical sheet materials which are in common use, and Fig. 1 gives
the B-H curves of the steels most usually employed.

The choice of one or other of these materials for any given equipment is usually well founded. Consider, for instance, rotating electrical machines. The aim of the designer is to crowd on to a given periphery that combination of maximum flux and ampere wires which will give the greatest output for the particular frame size. The periphery is for this reason divided up into slots for copper and teeth for flux,
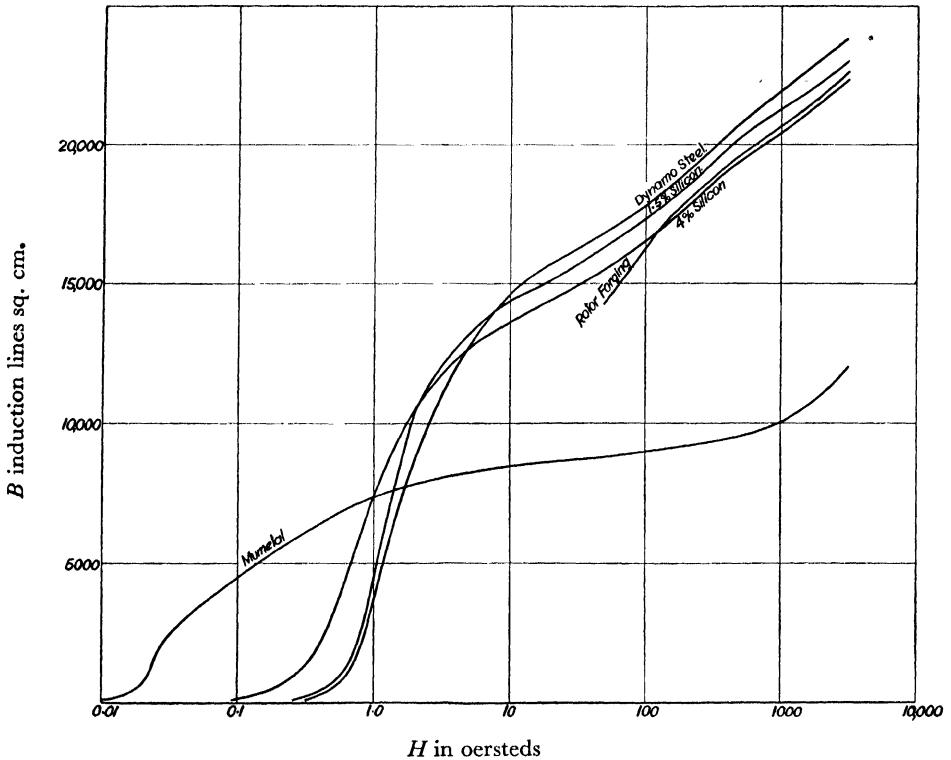

Fig. 1. *B-H* curves of steels commonly used.

remembering always that certain conditions must be met with regard to performance, depending on the type of machine. Chief amongst these, and common to all types of machines, is the limit of temperature rise which may be allowed without damaging the insulation of the conductors. Apart from the thermal limit there are usually a number of conditions imposed which relate to the electrical performance. In induction motors, for instance, the power factor of the motor must be kept reasonably high. The excitation of the machine is taken from the supply network, and excepting for iron losses represents so much wattless power, to which there is grave objection from the point of view of transmission and distribution of

electrical power, since it increases expenditure on plant and cables. Induction motors are therefore designed with the shortest air gap consistent with mechanical clearance, in order to reduce the wattless power required for excitation. In the effort to increase the output of the motor, tooth densities are increased until they begin to absorb an appreciable amount of the excitation ampere turns, and in turn tooth widths are increased until they encroach on the copper space to the point where further encroachment would cause difficulties due to the increase in the copper loss of the machine. In practice tooth densities approach 21,000 gauss in the narrowest sections, and to enable this to be done steel of the highest saturation intensity must be used; this is the low carbon sheet, usually designated "Dynamo Steel" (Fig. 1). If higher permeability at high densities could be obtained in an improved material it would be immediately utilized to increase induction motor ratings.

To turn now to D.C. machines, the necessity for reducing the exciting ampere turns is not so pronounced as in A.C. motors, since no question of loading the mains with reactive power arises. On the contrary, it is essential to ensure that the machine flux is maintained on load, and for this reason relatively long air gaps are used, so that the demagnetizing effect of armature current is not excessive. Magnetic saturation in the teeth reduces the area of the flux wave when distorted by the armature load ampere turns, and therefore the output of the machine. An increase in permeability in the region of high tooth densities (again about 21,000 gauss) would be of considerable help in increasing ratings from this point of view. If, for instance, tooth densities could be increased 10 per cent without increasing the ampere turns required by the teeth, then the axial length of the machine could be cut by the same percentage, and there would result a considerable saving of space and material. The air-gap length would be reduced at the same time so that the ratio of field to armature ampere turns and therefore machine stability would remain unaltered.

There is of course the difficulty that increased iron densities might result in increased losses, so that full advantage could not be taken of such an improvement in permeability, but it is important to realize that so-called stray losses are of overriding importance in highly rated machines, and these are very frequently a function of magnetizing force, rather than induction. For instance, field form entry loss in slot copper is proportional to the square of the M.M.F.

acting on the teeth, so that it would be reduced by the use of steel of higher permeability.

It can therefore be said that the barrier to increased ratings of A.C. and D.C. motors of given frame size is the limited value of the saturation intensity of the steels available. Steel with lower losses under alternating magnetization would of course assist in increasing ratings, especially in high-speed machines. In fact, it is common practice in many high-speed industrial D.C. motors to use $1\frac{1}{2}$ per cent silicon steel instead of dynamo sheet in order to improve efficiency, in spite of the reduction in saturation value (Fig. 1), but such objection to losses would be far outbalanced by any marked improvement in permeability.

With regard to yoke densities and back of core densities in D.C. machines, it is necessary to avoid as far as possible saturation effects in the interpole magnetic circuit, for reasons connected with commutation. These densities are therefore limited to the region of the knee of the magnetization curves of the materials used. Here again, then, an obvious advantage would result from materials with increased permeability in the higher density regions.

There is a further type of rotating electrical machine which deserves special mention, namely, the turbo-alternator. Such a machine consists of a high-speed solid rotor revolving in a laminated stator. The rotor is a slotted forging, the slots carrying the exciting winding of the machines. The limiting feature of high-speed rotor design is the mechanical strength of the rotor material, which must withstand the stresses set up by the centrifugal forces acting on the rotor winding. Rotor diameters are consequently severely limited in dimensions, and the available material must be loaded with as much flux as possible in the effort to increase the rating. A further requirement of rotor material is thus a high saturation value. The materials used are usually nickel steels, or nickel chromium steels, annealed before use, and the greatest care is exercised in examining the forgings for defects. Fortunately, the steels which give the best tensile strengths, when in the form of large forgings, possess a fairly high saturation value. This does not mean, however, that an increased saturation value would not be an advantage. The point is that under no circumstances could tensile strength be sacrificed for higher saturation. In the last twelve years tensile strengths of large forgings have been increased from 36 to 48 tons per sq. in. without any appreciable reduction in saturation value. On the stator the position magnetically

is eased due to increasing diameter, and limitations in permeability and saturation values of the materials used for stator construction are not a vital factor. On the other hand, losses become exceedingly important in large machines. The efficiency of a modern 50,000 kw. generator is about 98 per cent, i.e. a total loss of 1000 kw., of which iron loss might represent about 250 kw. Although the percentage loss is remarkably low, the difficulties of dissipating so many kilowatts of heat energy cause elaboration in design and increase in size and cost. For instance, the radial core ventilating ducts of a large machine increase its length by as much as 20 per cent. Anything that can be done to reduce the loss is thus of extreme importance. Low-loss steel, usually with $3\frac{1}{2}$ or 4 per cent silicon content, is therefore used for stator construction. The high electrical resistivity of this steel assists very materially in reducing iron losses due to eddy currents set up by fringing fluxes.

Turning now to power transformers, for which larger quantities of magnetic sheet are used than for any other type of equipment, the important properties of the steel are losses and permeability over a range of densities considerably lower than those used in rotating machines.

Transformers are normally continuously energized. The waste of energy due to iron losses in all the transformers of a supply network may reach 6–8 per cent of the total energy delivered, bearing in mind that between generator and consumer three or more transformations may take place. Fig. 2 shows the percentage iron losses to be expected with varying size of transformer and the annual cost of the iron losses as a ratio of the first cost of the transformer. The capitalized cost of iron loss may greatly exceed the bare cost of the transformer, so that the importance of using low-loss steel is at once apparent. Consequently 4 per cent silicon steel is in almost universal use for transformer cores. The working densities are also limited, partly because of this necessity for reducing loss, but also for additional reasons connected with the magnetizing kva. required. The percentage magnetizing current for varying size of transformer is shown in Fig. 2. The reason for limiting magnetizing current has already been referred to in connexion with induction motors. There is, however, a further aspect of importance in transformer design, namely, the distortion of the magnetizing current from sinusoidal form due to the non-linearity of the B-H curve for the steel. This distortion results in magnetizing currents with a large harmonic content. Fig. 3 shows the
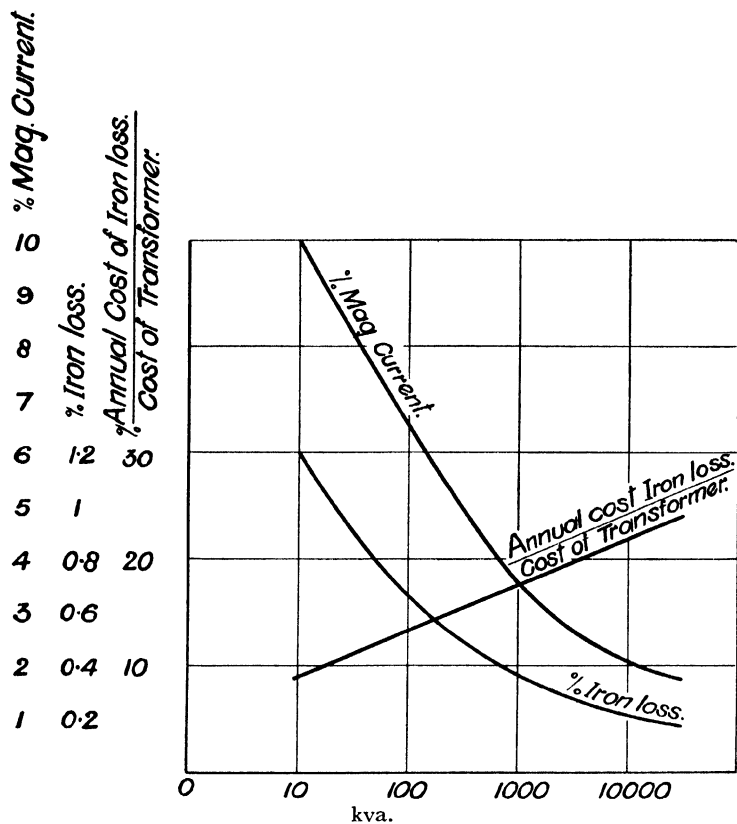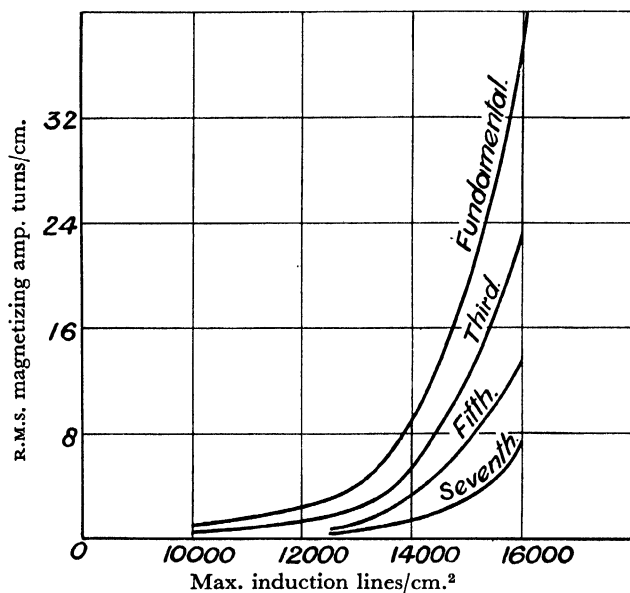
Fig. 2. Transformer losses.

Fig. 3. Harmonic components of magnetizing current
for 4 per cent silicon steel.

amount of such harmonics for transformer sheet magnetized with sinusoidal flux. Harmonic currents in power networks are very undesirable for a variety of reasons. They lead to increased interference with communication networks, increased losses in generating equipment, commutation difficulties in converting machinery, and regulation difficulties, especially on long lines. It is probably more for these reasons than any other that transformer flux densities in this country are limited to about 13,500 gauss, and in many cases much lower densities are used. The magnetizing force required by transformer steel at 13,500 gauss is usually about 10 oersted. If for the same force steel were available giving a density of say 17,000 gauss, the working density could be increased to this figure, and a very vital increase in output for a given frame size would result. The position at the moment is that whilst reduction in losses is very desirable, such reduction would be of very much enhanced value if it were accompanied by an increased permeability in the region where H is 10 oersted.

The present discussion on sheet materials would not be complete without some reference to the mechanical properties of electrical sheet steels, as far as they affect production and design. The most vital aspect is that of the space factor obtainable in a built-up core, which depends on the uniformity of thickness and the surface finish of the laminations. It is gratifying to be able to record that present-day mill practice has achieved very marked improvement in both these properties in recent years. A designer must allow sufficient margin to cover the worst space factor likely to occur, so that variation in space factor leads to wasteful design. Variation in thickness leads in addition to slack cores and subsequent vibration and noise. Finally, there is the trouble due to the existence of rolling stresses in sheets, which cause punchings cut from the sheets to buckle and bend. Besides leading to constructional difficulties, such defects lead to bending stresses in the core plates, which in turn result in increased loss.

Before concluding it may be of interest to review recent trends in the improvement of electrical sheet steels. In America there is a tendency to use higher percentage silicon steels, in the effort to reduce losses. 5 per cent silicon sheet is used in fairly large quantities, and there has been some attempt to use 6 per cent silicon sheet. The increased brittleness which results from the higher silicon content is a deterrent against its greater use. Indeed, in order to handle it

successfully a special shop technique is necessary, in which the sheets are cut before annealing, and then hydrogen annealed in the factory. Excepting for the largest scale manufacture, it appears that such methods would not be economical, and are therefore unlikely to gain much ground in this country. Moreover, there is the additional viewpoint, that a reduction in permeability resulting from the increased silicon content occurs in the density regions where a higher permeability is most urgently required.

A further development is the adoption of a combination of cold working and thermal treatments in the production of steels to give a greater preferred orientation of the crystal alignment in the sheets than has hitherto been obtainable in hot-rolled sheets. Very promising results have been achieved already, and a considerable increase in permeability and reduction in loss have been obtained when the direction of magnetization is along the rolling direction, though, it should be added, at the expense of properties transverse to this direction. At the moment it appears that the development may have important effects on transformer construction, but not on machines.

Finally, there is the production of nickel-iron alloys with remarkable magnetic properties. It must, however, be said here, that whilst they have been of immense value in the design of special apparatus such as current transformers, communication and other high-frequency equipments, they have had little or no influence on the design of power plant. The reason is of course that they invariably suffer from a low saturation value, and are therefore of no use in the effort to raise the magnetic loading of material.

In conclusion, it is hoped that this brief review will have demonstrated that there are many cases in practice in which the properties of available magnetic materials impose restrictions on the designer, and where improved magnetic properties could be immediately utilized to increase ratings, or to give better performance, objects which the successful engineer must ever keep before him.

Lecture IV

MAGNETIZATION CURVES OF FERROMAGNETICS

BY

EDMUND C. STONER, Ph.D., F.Inst.P., F.R.S.

Reader in Physics at the University of Leeds

Lecture IV

INTRODUCTION

THE peculiar character of the variation with field of the magnetization of ferromagnetics has long attracted attention, and, partly on account of their technological importance, the forms of the magnetization curves have been extensively investigated. The earlier attempts at explanation of these curves, though often full of interest and superficially successful, were usually based on simplifying assumptions which have since proved untenable.

The essential distinctive property of ferromagnetics is that they acquire strong magnetization in relatively weak fields. This is not because the elementary magnets (associated with the atoms or electrons) have unusually large moments, but because of the special character of the interaction between them, and this has now received a satisfactory interpretation within the general quantum theoretical scheme. The basic theory of the nature of ferromagnetism, however, does little towards the interpretation of the form of the magnetization curves, which depends in a complicated way on a number of secondary effects. Although it is not yet possible to give a detailed quantitative explanation of the precise forms of the magnetization curves of particular materials, some of the main determinative factors are now clear, and it is with a discussion of these that this paper is mainly concerned. The properties of particular materials are considered more fully in other papers of this series.

In order that the effects to be discussed may be seen in their proper perspective, it is perhaps desirable to refer first, very briefly, to a few salient points in the general theory. In the correlation of ferromagnetic phenomena the value of the molecular field hypothesis of Weiss has long been recognized. After Langevin had developed his theory of paramagnetism (1905), Weiss extended the treatment (1907) and investigated the effect of an internal quasi-magnetic field proportional to the intensity of magnetization. He showed that such a field would result in a spontaneous magnetization which decreased more and more rapidly with increasing temperature to become zero at a critical temperature, the Curie temperature, θ. At higher temperatures, the theoretical substance would behave as a paramagnetic, with a susceptibility inversely proportional to the excess of

the temperature above θ. In order that the theory should be applicable to actual ferromagnetics, a further supposition was necessary. Ferromagnetics are not necessarily magnetized in the absence of an external field, and for this fact to be compatible with the theory it is necessary to suppose that the spontaneous magnetization, while practically constant in magnitude throughout the material, may vary in direction, being uniform in direction (when the bulk magnetization is zero) only throughout small regions, generally called domains. The effect of an external field is to align the directions of magnetization of the domains, so that the ordinarily observed saturation magnetization is a fair measure of the intrinsic spontaneous magnetization.

When the two hypotheses of the Weiss theory are combined—that of a molecular field proportional to the magnetization, and of a domain structure—a remarkable degree of co-ordination becomes possible, not only of many of the magnetic, but also of the characteristic thermal properties of ferromagnetics. The molecular field which must be postulated is, however, thousands of times greater than that which would arise from purely magnetic interaction, and its origin remained completely obscure until Heisenberg (1928) showed that effective fields of the right order of magnitude could arise from quantum mechanical exchange effects; these are of the same kind as are involved in homopolar molecule formation. It had become clear, before this development, that the effective magnetic elements in ferromagnetics were electron spins, the most direct evidence being provided from experiments on the gyromagnetic effect. The exchange interaction in ferromagnetics must be such as to favour parallel alignment of the electron spins rather than anti-parallel, as is usual in simple molecules. The detailed theory of the exchange interaction effects is complicated, but considerable progress has recently been made by attacking the problem of ferromagnetism on a collective electron basis, which has proved so valuable in the theory of metals generally, rather than by Heisenberg's original method. For the present purpose, however, the developments in connexion with the fundamental theory need not be considered further.

To summarize, ferromagnetics may be regarded as constituted of spontaneously magnetized domains in which there is alignment of electron spins. The process of magnetization consists in the changing of the direction of magnetization of the domains towards the field direction, the ultimate magnetic elements being electron spins. It is

with this basic picture in mind that the attempt to interpret magnetization curves must be made. The question of the status of domains is one which is most conveniently taken up after fuller consideration of the magnetization process.

The general character of magnetization curves is well known. If a gradually increasing field is applied to an initially demagnetized ferromagnetic, the magnetization at first increases slowly and approximately reversibly; the initial slow increase is followed by a more rapid and largely irreversible increase; and finally there is a slow and approximately reversible increase towards the saturation value. The subdivision of the process into three stages is convenient, although ordinarily these stages are not sharply distinct. The general interpretation, to be considered more fully, is that the first stage corresponds to reversible shifts of the boundaries of regions of unidirectional magnetization, that is, of domain boundaries; the second to irreversible jumps of the directions of magnetization of the domains through large angles, usually $180°$ or $90°$ (or, which comes to the same thing, to spontaneous irreversible shifts of domain boundaries); and the third to reversible turning of the directions of magnetization towards that of the field. The general character of the hysteresis curve traced out when a cyclic variation of the field is made does not require description. A satisfactory explanation of the form of the initial curve should clearly embody the means of interpreting the complete hysteresis curve.

In detail, the magnetization curves differ greatly not only for chemically different materials but also for chemically similar materials which have undergone different thermal and mechanical treatment, and the problem at issue is to account, at least qualitatively, for these differences. It is the properties of ordinary polycrystalline material which are of the greatest technical importance, but the general problem is best approached by a consideration of the magnetic characteristics of single crystals; for the properties of a polycrystalline specimen must depend on those of the constituent crystal grains.

SINGLE CRYSTALS

Magnetization. A typical set of magnetization curves for a single crystal of iron is shown in Fig. 1.

Iron has a body-centred cubic structure. For the field in the direction of a cube edge (100), the intensity of magnetization rises in weak fields practically to the saturation value, I_0. (The magnetization

in very weak fields will not be discussed, partly because individual crystals differ widely, depending on their degree of perfection, and partly because, for the general problem, it is the behaviour of crystals in moderate fields which is most significant.) Along the face diagonal (110) the intensity rises rapidly to approximately $I_o/\sqrt{2}$, and along the cube diagonal to $I_c/\sqrt{3}$, when there is a fairly well-defined bend in the curves, the magnetization subsequently increasing gradually to the saturation value. The supposition that the spontaneous magnetiza-
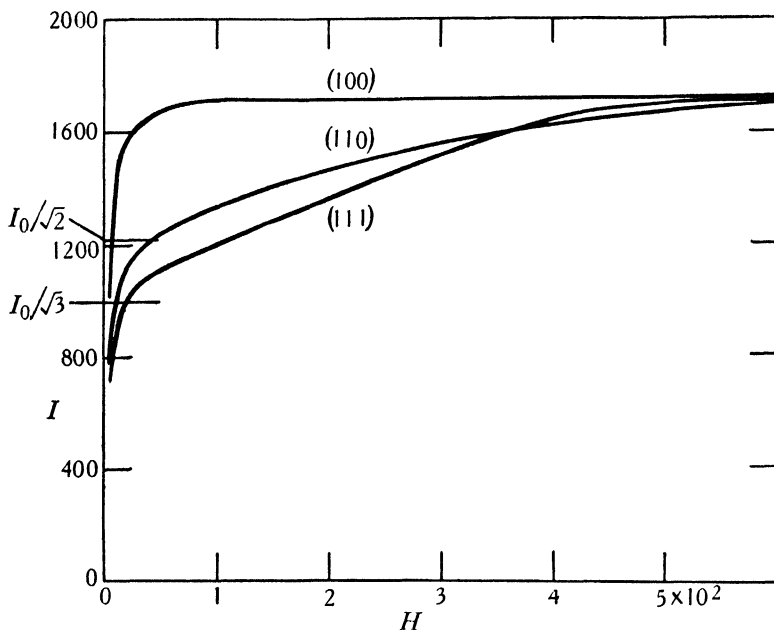

Fig. 1. Variation of magnetization (I) with field (H) in the 110 plane of iron. The 100, 110 and 111 directions correspond to the cube edge, face diagonal and cube diagonal. (Honda and Kaya, 1926.)

tion is unidirectional only over restricted regions (domains), in the absence of a field, is as necessary for single crystals as for poly-crystalline material; for even a single crystal is not ordinarily spontaneously magnetized to any considerable degree as a whole. The general character of the magnetization curves then receives an immediate explanation if it is assumed that in the absence of a field the domain directions of magnetization are distributed at random along the six equivalent cubic axis directions, these being directions of "easy" magnetization.

A comparatively weak field is sufficient to change the direction of magnetization in a domain from its original easy axis to the easy axis

(or axes) making the smallest angle with the field. If the 100 axes are chosen as X, Y, Z co-ordinates (Fig. 2), for a field along a face diagonal (say in a positive direction in the XY plane) half the domains will then be magnetized along the positive X, half along the positive Y direction, giving a resultant magnetization in the direction of the field of $I_0/\sqrt{2}$, the value at which the bend in the magnetization curve is observed.

A similar argument accounts for the position of the bend in the 111 curve. When the field increases above the small value necessary to give the maximum resultant magnetization consistent with the directions of magnetization of the domains remaining along easy axes, the magnetization can increase further only through the
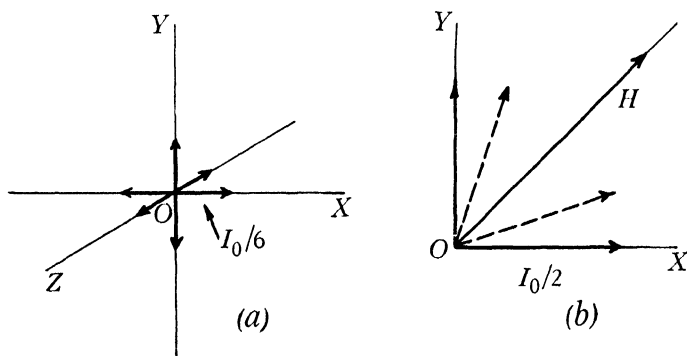

Fig. 2. Distribution of directions of magnetization (a) in demagnetized state and (b) in weak fields (full arrows) and moderate fields (dotted arrows) along face diagonal, for iron.

magnetization direction in the domains leaving the "easy" axes and turning towards the field (as suggested by the dotted arrows in Fig. 2). An immediate measure of the energy required for the completion of this process is given by the area between the I axis and the magnetization curve (i.e. $\int_0^{I_o} H\,dI$).

The behaviour of a crystal may be formally described by giving the values of anisotropy coefficients in the general expression for the energy, as dependent on the direction of magnetization relative to the crystal axes. Let U_o be the energy per unit volume of a crystal magnetized along one of the cubic axes; then the energy expression when I_o has direction cosines α_1, α_2, α_3 relative to the cubic axis co-ordinate system, retaining only the lowest term in the α's, may be written in the form

$$U\left(\alpha_1, \alpha_2, \alpha_3\right) = U_o + K\left(\alpha_1^2\alpha_2^2 + \alpha_2^2\alpha_3^2 + \alpha_3^2\alpha_1^2\right). \tag{1}$$

This gives, for example, for the excess of energy required to magnetize a crystal along a cube diagonal, (111) $(\alpha_1 = \alpha_2 = \alpha_3 = 1/\sqrt{3})$ and a face diagonal, (110) $(\alpha_1 = \alpha_2 = 1/\sqrt{2}; \alpha_3 = 0)$,

$$U_{111} - U_{100} = K/3, \\ U_{110} - U_{100} = K/4. \tag{2}$$

Equations of this type provide one means of determining K from the experimental data. The actual form of the energy expression is determinable purely from symmetry considerations—in this case, that it must be compatible with cubic symmetry. The expression given is adequate for nickel, but for iron agreement with experiment is somewhat improved by including a further term of the type $K' \alpha_1^2 \alpha_2^2 \alpha_3^2$. By applying the condition that the total free energy (that is, the internal energy given by the above expression together with the external field energy, $-HI \cos \theta$, θ being the angle between H and I) is a minimum, the forms of the magnetization curves above the bend may be calculated, and also the direction of magnetization for any direction of the field relative to the crystal axes. The observational results are well reproduced. This means, not that the properties of the crystal are "explained", but that they are satisfactorily summarized by means of the anisotropy coefficients. The atomic theory problem then becomes that of accounting for the magnitude of these coefficients. Qualitatively the theory is here satisfactory in that it shows that anisotropy effects of the right order of magnitude can arise as a result of the essentially magnetic interactions between spin and spin, and spin and orbital moment of the electrons.

For iron K is positive, corresponding to the cube axes being easy directions. For nickel K is negative, and numerically some ten times smaller, the "easy" direction in nickel (face-centred) being the cube diagonal. For cobalt (hexagonal) the situation is simpler, as there is a single well-marked easy direction along the hexagonal axis. The excess energy expression takes the form

$$U(\phi) = U_o + K \sin^2 \phi + K' \sin^4 \phi, \tag{3}$$

where ϕ is the angle between I_o and the hexagonal axis.

The curves are illustrated in Fig. 3. Numerically K is about ten times as large as for iron, corresponding to the larger fields required to saturate cobalt in other than the easy direction.

Magnetostriction. As illustrative of results on magnetostriction, those of Webster for iron are shown in Fig. 4. Whereas iron expands

for magnetization along a cubic axis, nickel and cobalt both contract for all directions of magnetization.

The change of length of a single crystal on magnetization is not,
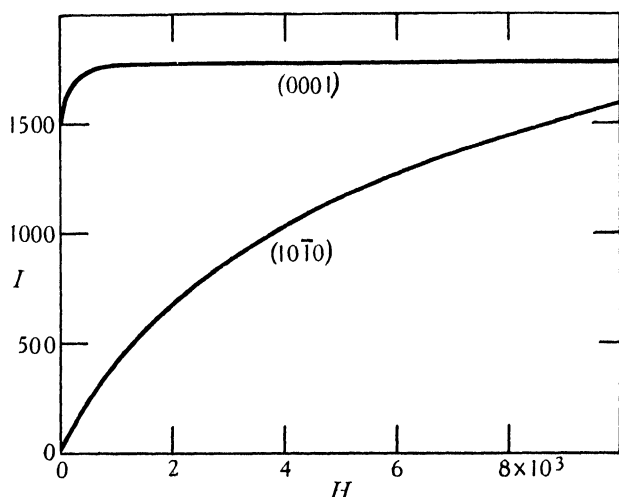

Fig. 3. Magnetization curves for cobalt single crystal along hexagonal axis (0001), and normal to hexagonal axis (10$\bar{1}$0). (Kaya, 1928; Honda and Masumoto, 1931.)
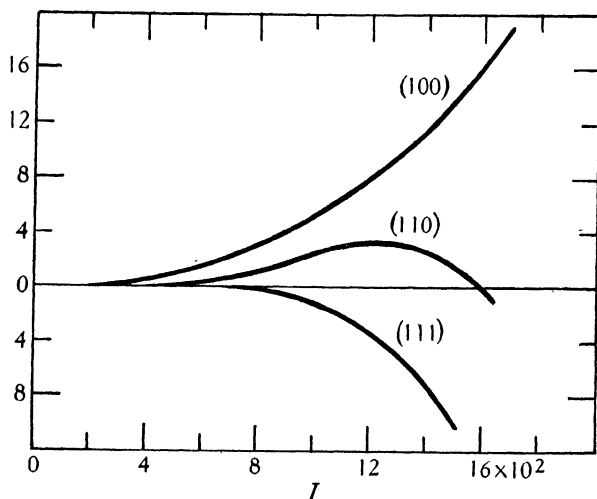

Fig. 4. Relative change of length with intensity of magnetization in a longitudinal field along the cube edge (100), face diagonal (110) and cube diagonal (111) directions for iron single crystals. (Webster, 1925.)

perhaps, in itself, of particular interest. The single crystal curves do, of course, bring out peculiarities which are masked in the curves for polycrystalline material; but, beyond this, the results are of great

importance in connexion with the general interpretation of magneti-
zation curves, and for that reason must be further considered. A
formal co-ordination of the results is obtained by an extension of the
treatment indicated above. A more general expression for the energy
of a crystal will include terms corresponding to the deformation
energy, involving on the one hand purely elastic coefficients, and
on the other coefficients which may be termed magneto-elastic,
arising jointly from the magnetization and the deformation. Of these
last the important coefficients, for a cubic crystal, are λ_{100} and λ_{111},
which are physically the relative changes in length for magnetization
to saturation along a cube edge and a cube diagonal, immediately
obtainable from the experimental results. The form of the complete
energy expression for a magnetized deformed crystal is again deter-
minable from symmetry conditions; this energy expression, involving
the experimentally determinable K, K', λ_{100}, λ_{111} and the appropriate
elasticity coefficients, then embodies a description of a whole range of
magnetic and magneto-elastic properties of the crystal. It is un-
necessary to discuss the treatment in detail here—it will be sufficient
to refer to a few general and simple points. It is easy to show, by
straightforward energy considerations, that there is a relation between
the magnetostriction and the effect of tension on magnetization, a
relation which may be put in the form

$$\partial\lambda/\partial H = \partial I/\partial Z, \qquad (4)$$

where $\lambda = \Delta l/l$ and Z is the applied tension. This relation at once
shows that the effect of tension is to increase magnetization if λ is
positive and vice versa. The effect of stress on a crystal is essentially to
modify its effective symmetry, and so the character of its magnetic
anisotropy. In particular, if a cubic crystal with easy directions along
the cubic axes and with positive λ is stretched along one of the cubic
axes, the direction of stretching becomes an energetically favoured
easy direction; in contrast, if λ is negative, the direction at right
angles is favoured. The importance of these ideas lies rather in their
application to polycrystalline material; for if the applied stress is
great enough it may be sufficient to mask almost completely the
natural anisotropy effects in the individual grains, and to produce a
quasi-crystalline behaviour. The order of magnitude of Z for the
"artificial" anisotropy effects to become comparable with the
natural effects is

$$Z \sim K/3\lambda. \qquad (5)$$

This expression indicates values of the order 4 kg. per sq. mm. for nickel, and 80 kg. per sq. mm. for iron.

No reference has been made to the atomic theory of magnetostriction, for, although it seems fairly clear what are the main factors to be considered, reasonable quantitative agreement with observation has not been obtained. This is undoubtedly due in part to the complexity of the problem, but the position is hardly satisfactory in that the reason for the opposite sign of the magnetostriction in iron and nickel is still obscure.

This survey of the properties of single crystals may appropriately be concluded with a table of the values of the coefficients which have been discussed, and some other data.

Table I. Crystal constants for iron, nickel and cobalt

	I_0	$K \times 10^{-5}$ (erg/cm.^{3})	$K' \times 10^{-5}$	$\lambda_{100} \times 10^5$	$\lambda_{111} \times 10^5$
Fe	1716	4·0 to 4·8	∼ 3	1·8	−1·3
Ni	508	−0·4 to −0·6	∼ 0	−5·2	−2·8
Co	1422	41 to 44	∼10	—	—

POLYCRYSTALLINE MATERIAL

Approach to saturation. In dealing with polycrystalline material it is a natural first step to consider what would be the properties of an aggregate of crystals, orientated at random, each having the somewhat idealized characteristics which have been discussed. By the appropriate averaging process, Gans (1932) has obtained the following expressions:

For weak fields:
$$I = I_0 \{A + B \ (I_0/K) \ H \ ...\};\tag{6}$$

For strong fields:
$$I = I_0 \{1 - \alpha \ (K^2/I_0^2) \ (1/H^2) \ ...\}.\tag{7}$$

In these expressions A, B, α are calculable constants depending on the directions of easy magnetization in the crystal. For iron, for example, $A = 0·831$, $B = 0·150$, $\alpha = 0·0872$. The weak field expression is not directly of much interest, for it is based on the assumption that the directions of magnetization swing round to the easy axis directions nearest to the field in vanishingly small fields, so that the complex low field behaviour of most polycrystalline specimens is outside its range of application. The strong field expression indicates that for I/I_0 approaching unity, a given value of I/I_0 will be reached at a value of

H proportional to K/I_o. For $I/I_o = 0.95$, for example, using the mean values from Table I, the value of H for iron is about 330. (The corresponding value for nickel is $H = 120$.)

The strong field expression, which is of the form

$$I = I_o \{1 - b/H^2 \dots\}, \tag{8}$$

in some cases reproduces the experimental curves very closely, as for example for well-annealed specimens of electrolytic iron. A calculation of the anisotropy constant K from the experimental b gave a value agreeing well with that found directly with single crystals. It is of interest that in this way a single crystal characteristic constant can be determined from observations on polycrystalline material. In general, however, the agreement with equation (7) is not close, though sometimes the general form of equation (8) is a fair approximation. An essential assumption involved in equation (7) is that the individual crystals are unstrained. Internal strains will have the effect of modifying the effective anisotropy constants of the crystals. Although the approach to saturation would still be represented by an equation of the form of (8), with large internal strains the coefficient b would be determined by the strain anisotropy rather than the natural anisotropy. The manner of approach to saturation thus gives a measure of the internal strain. The greater the anisotropy, the greater b, which ordinarily has a minimum value when the crystals are unstrained; from the excess of b above the value indicated by equation (7) an estimate of the degree of internal strain may be obtained.

For a considerable number of materials (for example "permalloy" in fields up to about $H = 2000$, and nickel in higher fields) the approach to saturation is approximately represented by

$$I = I_o (1 - a/H \dots). \tag{9}$$

A process giving this dependence on H is the reversal of small domains (that is the reversal of the groups of spins in these domains). Although the jump processes (to be discussed below) for domains constituting the greater part of the material occur in fairly low fields, there is a calculable probability of an appreciable number of small domains remaining with magnetization opposite to the direction of the field until the field is much larger; the jump processes in these small domains may then be responsible for the $1/H$ approach to saturation. The point to be emphasized is that empirical approach laws differing from equation (8) are in many cases required by the observations, and indicate the occurrence of other processes than

those taken into account in the derivation of equation (7); there is, however, little doubt that the main factor determining the manner of approach to saturation for most materials in the field range of technical importance is the natural and strain anisotropy effects in the crystal grains.

The characteristic distinguishing the processes which have been considered as mainly involved in the region of approach to saturation
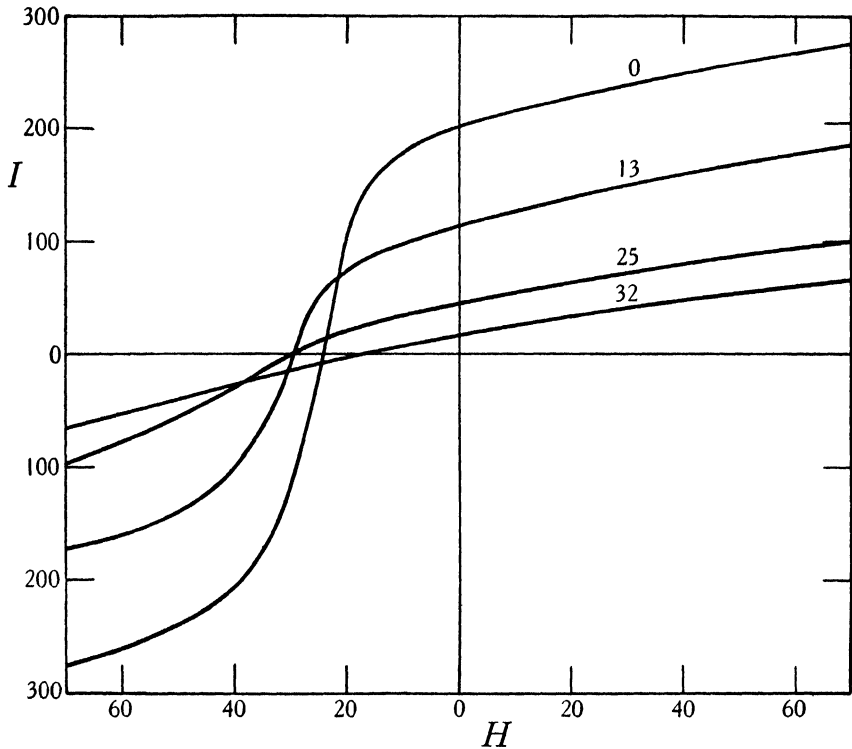

Fig. 5. Effect of tension on the magnetization curves of material with negative magnetostriction. Hard drawn nickel. The numbers on the curves give the tension in kg. per sq. mm. (Becker and Kersten, 1930.)

is their reversibility. Usually for polycrystalline material reversible changes account for only a relatively small part of the total change in magnetization. In homogeneously strained material, however, practically the whole change may be reversible. This occurs for materials with negative magnetostriction under sufficiently high tension. The magnetization curve of a nickel wire under increasing tension approaches a form similar to that for a cobalt crystal in the difficult direction (Fig. 3). The effect is illustrated in the curves in Fig. 5.

When the tension is sufficiently large, the "easy" directions (and so the directions of spontaneous magnetization of the domains) are at right angles to the axis of the wire, and the change in magnetization in a longitudinal field occurs entirely by the turning process. The polycrystalline wire behaves as a pseudo-crystal, with λZ as the effective anisotropy coefficient, and the approach to saturation region extends over the entire magnetization curve. There is a similar effect in material with positive magnetostriction subjected to compression. This is well shown in the curves given in Ewing's *Magnetic Induction in Iron and other Metals* (1900), as well as in more recent work.

Irreversible processes. On the steep part of a magnetization curve the greater part of the change in magnetization is irreversible, as has long been known. It is only fairly recently, however, that it has been found that the changes are discontinuous. In Barkhausen's experiment (1919) the specimen is surrounded by a search coil connected through an amplifier to a telephone. On varying the field gradually (say by bringing up a magnet, or varying the current in a field coil by means of a liquid resistance) a rustling is heard; indicating that the electron spins change direction in groups large enough for the change in B to produce an audible effect. The rustling sound results from a succession of signals from the groups. Refinements of the original experiment, notably by Bozorth and his collaborators, have enabled estimates to be made of the size of the individual groups. A typical set of results is shown in Fig. 6.

The average size of the groups in which the electron spins reverse suddenly rises to a maximum at or near the steepest part of the magnetization curve. A limit is set to the size of the group which can be detected by the sensitivity of the apparatus. It cannot therefore be concluded that no group reversals occur in the initial and final stages of magnetization, but merely that the groups must then be much smaller than those whose reversal accounts for practically the whole change on the steeper parts of the magnetization curves. Here only a very small fraction of the change is due to reversals of groups containing less than about 10^{10} atoms (corresponding to a volume of 10^{-13} cm.^{3}). A striking and rather peculiar result of Bozorth's investigations is that the maximum "average size" of the groups (the average size varies over the magnetization curve as shown in Fig. 6) does not differ greatly for different materials. Values ranging from about 10^{-9} cm.^{3} for a specimen of iron to about 5×10^{-8} cm.^{3} for a 50 per cent ferronickel were found, corresponding to groups of

some 10^{15} atoms. Groups of the same order of size were found for annealed and hard-worked specimens and for single crystals. The important general results are then twofold—first, that the size of the groups varies over a wide range in any individual specimen (that is, there is no indication of a particular "block" size), and secondly, that for the different materials the average size is much the same (that is, there is no indication of a well-defined dependence on grain size). In hard-worked material the grain size may be smaller than
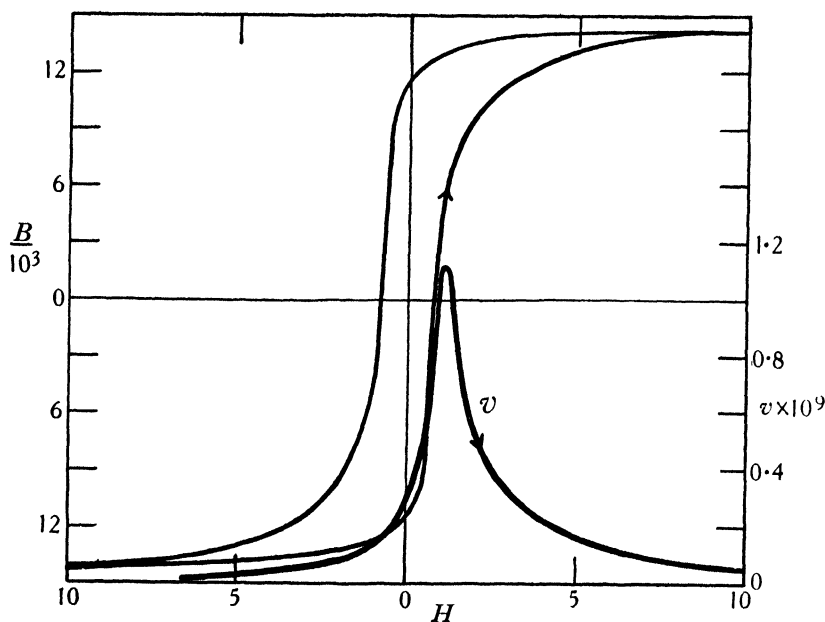

Fig. 6. Hysteresis loop and v-curve for annealed iron. $v =$ average volume in which sudden reversals of magnetization occur. (Bozorth and Dillinger, 1930.)

that of the average Barkhausen group, but usually the groups are smaller than single grains.

Although the reversal process may be "sudden", it can hardly be instantaneous throughout a group. It was suggested by Langmuir that reversal might start at a "nucleus" and spread in a wave-like manner. In suitably strained material, as was first observed by Forrer (1926), large single Barkhausen jumps may be obtained, corresponding to a considerable fraction of the total change in magnetization from saturation in the positive to that in the negative direction. This occurs for materials with positive magnetostriction, notably ferronickels with less than 80 per cent nickel, under longi-

tudinal tension. An example is shown in Fig. 7, which may be compared with Fig. 5.

For a wire under increasing tension the hysteresis curves approach a simple rectangular form, the magnetization reversing throughout the wire when a critical value of the field is passed. An extensive study of these large Barkhausen jumps has been made by Sixtus and Tonks (1931 onwards), and by Preisach (1929 onwards). Starting with a wire magnetized to saturation (to the left, say) a region of reversed
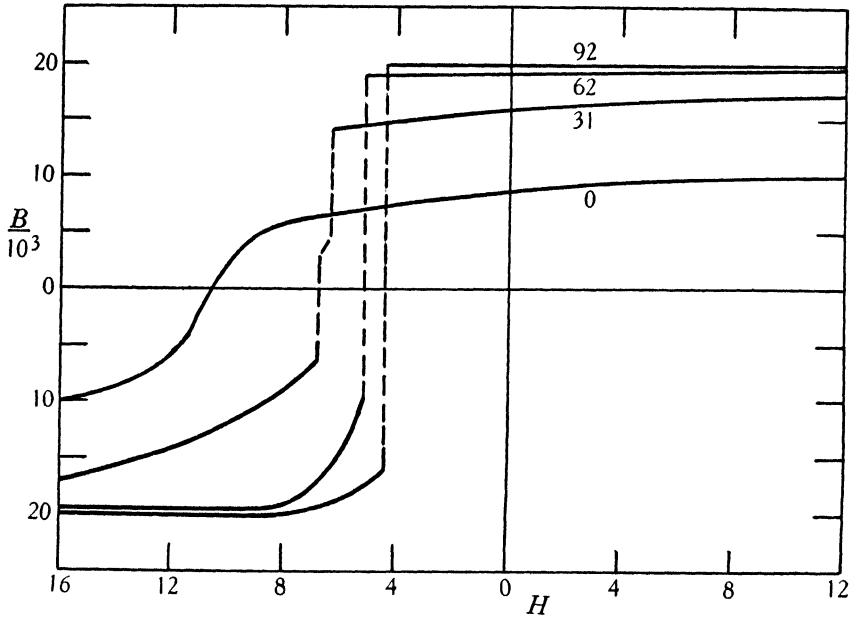

Fig. 7. Effect of tension on the hysteresis curves of material with positive magneto-striction. Ferronickel alloy, 15 per cent iron. The numbers on the curves give the tension in kg. per sq. mm. (Sixtus and Tonks, 1931.)

magnetization may be initiated by a localized "starting field", H_s, near one end. In effect the boundary of this region then travels (to the right) through the wire provided that the general applied field exceeds a critical value, H_o. (By very ingenious arrangements the velocity of propagation has been measured. It is approximately proportional to $H - H_o$. The process involves the dissipation of energy in the form of eddy currents, and the theoretical calculation of the velocity presents a complex problem which has not yet been com-pletely solved; this particular problem, however, need not be con-sidered here.) The values of the critical fields H_s and H_o depend on

the tension; as the tension is decreased both H_s and H_o approach a value approximating to the coercive field, H_c, for the unstrained material.

With the experimental arrangement adopted there is a well-marked "easy" direction for magnetization along the wire; for reversal to occur energy is required for the formation of a boundary between two regions of opposite magnetization, as discussed fully in the section below on the status of domains. Once a sufficiently extended boundary is formed, it travels "spontaneously" along the wire in the direction of the field; there is an increase in the resultant magnetization of the wire without an associated increase in the applied field. The process corresponds to a decrease in the total energy, due to the negative increase in the part dependent on the applied field. Owing to the "uniformization" of strain by the application of a large tension, the conditions are much simpler than in unstrained material, in which the localized internal strains have a dominating effect on the course of magnetization. The general picture obtained can, however, clearly be extended to the jump processes in the smaller groups constituting the domains of material under ordinary conditions. A Barkhausen jump may be said to correspond to a spontaneous irreversible shift of the boundary between two regions of differently directed magnetization; the factors determining the values of the field for which this occurs are most conveniently discussed after further consideration of the reversible processes which precede the jumps, and which predominate in the initial stages of magnetization.

Initial permeability. If a gradually increasing field is applied to an initially demagnetized specimen the increase in magnetization is at first reversible and approximately proportional to the field, though the range of reversibility and linearity differs widely for different materials. At any part of the magnetization curve the differential susceptibility $(\kappa_d = dI/dH; \ \mu = 1 + 4\pi\kappa)$ may be regarded as the sum of a reversible and irreversible susceptibility $(\kappa_r + \kappa_{ir} = \kappa_d)$, the value of κ_r being obtainable by superposing a small reverse or cyclic field on the strong field. The reversible susceptibility is very closely a function of the magnetization only, and has a maximum value for $I = 0$, this corresponding to the initial susceptibility κ_o.

The increase in magnetization due to rotation from easy axes towards the field against the crystalline field is far too small to account for the observed values of κ_o. The most promising interpretation is in

terms of a reversible shift of domain boundaries, leading in effect to
a growth of those domains magnetized in the field direction at the
expense of their neighbours. The occurrence of a domain structure is
undoubtedly conditioned by internal strains and, by considering a
very simple model, Becker (1932) has been able to show the kind of
relation to be expected between internal strain and initial suscepti-
bility. The random strains are idealized as arising from alternating
pressures and tensions in neighbouring regions. For a material for
which the magnetostriction is uniformly either positive or negative
this will give rise to alternate regions in which the magnetization, I_o,
is along and at right angles to the field.

The internal stress may be represented by
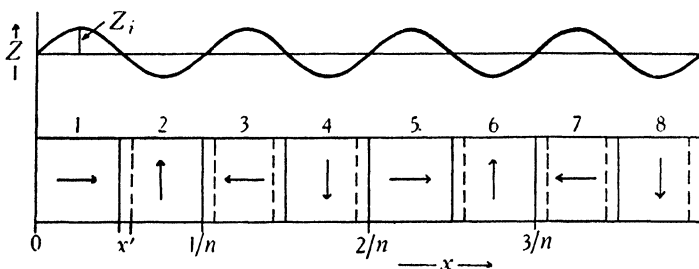
$$Z = Z_i \sin 2\pi nx, \tag{10}$$

Fig. 8. Directions of magnetization under alternating tension and compression.
The full and dotted lines indicate the positions of the boundaries in the
demagnetized state and in an applied field. (After Becker, 1932.)

where $2n$ is the number of boundaries per unit length. In the
demagnetized state the directions of magnetization will be distributed
as suggested by Fig. 8. When a field is applied the boundaries will
shift in such a direction as to increase the resultant magnetization.
A shift by an amount x' involves an increase in the magneto-elastic
energy (of amount $\lambda \int_0^{x'} Z \, dx$, where λ is the magnetostriction
coefficient) and a decrease of the external field energy (of amount
$-HI_o x'$). It follows that, for equilibrium,

$$Z = HI_o/\lambda, \tag{11}$$

and, provided that x' is small, the resultant magnetization is given by

$$I_H = HI_o^2/\pi\lambda Z_i, \tag{12}$$

corresponding to

$$\kappa_o = I_o^2/\pi\lambda Z_i. \tag{13}$$

Although it is obvious that the assumptions in this treatment are oversimplified for applicability to actual materials, it is equally obvious that they could be greatly modified in detail without changing the essential character of the result, which suggests at once, as conditions for high initial permeability, minimal internal strains and low magnetostriction.

Internal strains cannot be entirely eliminated, for in the most careful annealing of the purest material, magnetostrictive strains are set up as the material becomes spontaneously magnetized in cooling down from the Curie point. The magnitude of the internal strain Z_i arising in this way is of the order $E\lambda$, where E is Young's modulus; so that a theoretical upper limit to the initial susceptibility is given by

$$(\kappa_o)_{\text{max}} \fallingdotseq I_o^2/\pi\lambda^2 E. \tag{14}$$

This expression gives values in good agreement with the maximum values of κ_o for a whole series of iron-nickel alloys. It must be emphasized, however, that in any detailed treatment it would be necessary to take into account the usually different values of the magnetostriction in different directions. The formulae above can at best give indications of orders of magnitude.

An expression of the type of equation (13) is in general accordance with experience, in that materials in which, for other reasons, large internal strains are believed to be present have low initial susceptibility and vice versa, but in the absence of much more detailed information about the character and distribution of internal strains, it is hardly amenable to quantitative test. It is, however, also possible to make a rough estimate of the degree of internal strain from the form of the magnetization curve from saturation to remanence, as has already been stated, and the reasonable agreement between the estimates made by different methods provides evidence of the general validity of the treatment.

An important question arising from the consideration of reversible boundary shifts is as to the stage at which they will become irreversible. This will occur when a further virtual displacement is associated with an energy decrease. With the simple Becker model this would occur when the boundaries had shifted to regions corresponding to the maxima and minima of the stress curve, when the resultant magnetization would have the value $I_o/2$. This would imply values of the coercivity much higher than those ordinarily observed. An essential point which is rather obscured by Becker's treatment is

that the initial susceptibility depends inversely on the strain gradient at the boundaries, rather than on the maximum strain differences, whereas it is on these that the coercivity depends. More localized inhomogeneities of strain than those suggested by the sine wave form, separated by relatively large regions of approximately uniform strain, would result in the onset of the discontinuous process at a lower value of the resultant magnetization in the field direction, and the irreversible part of the process would account for a relatively larger part of the complete change in magnetization from zero to saturation. It is suggested that a more satisfactory interpretation of coercivity may be sought along these lines.

It should finally be emphasized that, in spite of the difficulties in detail in interpretation, both initial susceptibility and coercivity have been clearly and definitely linked up theoretically with the state of internal strain of the material, and that the present lack of quantitative precision in the treatment does not prevent its useful application in the practical consideration of the properties of particular materials.

THE STATUS OF DOMAINS

The concept of domains, in some form, plays such a prominent part in the interpretation of ferromagnetic phenomena that it is perhaps desirable to indicate briefly the evidence bearing on it and the extent to which it is amenable to more precise theoretical treatment.

As pointed out in the introductory section, the hypothesis of a domain structure is a necessary complement to that of a molecular field. That ferromagnetics are spontaneously magnetized, but, in the absence of an applied field, unidirectionally only over small regions, seems a straightforward deduction from the general form of the magnetization curves, and from the thermal and magneto-thermal properties of ferromagnetics, as shown by the excess specific heat, and by the magneto-caloric effect. The forms of the magnetization curves for a single crystal show at once that the extent of these regions cannot in general be identified with the volume of the crystal. The study of the Barkhausen effect has shown further that the volumes in which sudden jumps in the directions of magnetization occur are generally smaller than the grains in polycrystalline material, and usually contain some 10^{10} to 10^{15} atoms. The patterns formed by the deposition from suspension of colloidal ferromagnetic powders on polished single crystal surfaces (Bitter patterns) provide supplementary evi-

dence of some kind of "block" structure. These patterns often show remarkable regularities, and though their interpretation in detail is still problematical, the view seems inescapable that they arise in part from the existence of regions of uniform magnetization extending over many atoms, and yet small compared with the crystal size.

The magnetic evidence thus points clearly to the existence of a secondary structure in ferromagnetic crystals. It may be suggested, however, that this secondary structure is not of a kind peculiar to ferromagnetics, but that it is fundamentally similar in origin to the secondary structure in crystals generally, though it assumes a special character owing to the presence of those interchange interaction effects which give rise to ferromagnetism. Secondary structure generally is evidenced by the form of X-ray lines, by etch figures, by the occurrence of slip along a relatively small number of the planes which, in an ideal crystal, would be equivalent, and by the characteristic structures in many alloys indicating preferential precipitation along a limited number of planes. Domains are not explained in this way, but they become special exemplifications of a much more general phenomenon. On this view the domain question is of much wider significance than is generally supposed. The general problem, moreover, is one which it seems can be most hopefully attacked from the magnetic side, for the magnetization curves of ferromagnetics, both in their general form and their detailed peculiarities, are, to a large extent, sensitive indicators of the state of internal strain and irregularities of strain which are associated with a secondary structure. The problem in detail is complex, and the theory is far from being sufficiently developed for the extensive data available on the magnetic side to be utilized in full in the manner implied. It must be sufficient here to indicate the kind of relation which holds for the energy associated with the boundary between two regions of differently directed magnetization, which is fundamental for the whole problem.

The basic question is as to the amount of energy required for the breaking up of an ideal crystal magnetized to saturation in one direction into two regions magnetized in opposite directions. This question was first discussed by Bloch (1932), and since then his treatment has been somewhat extended and applied, notably by Becker. For simplicity it may be supposed that the crystal has a single "easy" axis of magnetization, and that the energy for magnetization at θ to this axis is proportional to $K \sin^2 \theta$. (The treatment would apply whether K is due to natural or to strain anisotropy.) The transition

between the states of magnetization to the left and to the right along the "easy" axis may be regarded as occurring by a rotation of the magnetization vector I_0 by an amount $\Delta\theta$ in passing from one layer of atoms to the next; the thickness of the transition region (the boundary) then being given by $\delta = (\pi/\Delta\theta)\,a$, where a is the distance between the atomic layers. The interaction energy between neighbouring layers (the most important part of the total interaction energy) is proportional to $J\cos\Delta\theta$, where J is the interaction integral, proportional to the interaction energy per atom. The excess energy (as compared with the state of uniform magnetization) is then proportional to $(\cos\Delta\theta - 1)$, or, since $\Delta\theta$ is small, to $(\Delta\theta)^2$; giving, for the whole transition region, an excess proportional to $J\Delta\theta$ or to J/δ. The interaction excess energy is thus greater the narrower the boundary. The excess due to the magnetic anisotropy is readily seen to be proportional to $K\delta$, that is, it is greater the wider the boundary. For minimum boundary energy it is then easily shown that the order of magnitude of δ is given by

$$\delta \sim a\,\sqrt{(J/K_a)}, \tag{15}$$

where J and K_a are the interaction and anisotropy coefficients per atom. Since J/K_a is usually of the order 1000, this indicates boundary widths of the order of 30 atoms. Further, the order of magnitude of the boundary energy per unit area is given by

$$E/A \sim (1/a^2)\,\sqrt{(JK_a)} \sim (\delta/a^3)\,K_a. \tag{16}$$

From the observed values of the anisotropy coefficients (the values in Table I must be divided by approximately 10^{23} to give the atomic coefficients) it is evident that the energy required for boundary formation is exceedingly small. Although the state of bulk magnetization to saturation is that of lowest energy for an ideal crystal under idealized conditions, even slight departures from idealized conditions would generally be sufficient, energetically, to favour the breaking up into domains; the inevitable demagnetizing field, favouring a state of zero bulk magnetization, is an example of such a departure. Moreover, the many ways of attainment of the domain structure weights the microscopically non-uniform states heavily as compared with the strictly uniform state, and will overwhelm the effect of the slight energy difference, as determinative of the normal state under ordinary temperature conditions. Most important, however, is perhaps the indication that even slight local inhomogeneities of

strain may be sufficient to favour the formation of magnetic domain boundaries, and to stabilize the magnetic domain structure. The rather specialized discussion given here must, of course, be considered in relation to the discussions of particular aspects of the magnetization curves in the earlier sections, where the general idea of domains has been extensively applied.

This short survey of the domain problem must necessarily be made up rather of suggestions of possibilities than of records of achievements. The main point to be emphasized is that the view that ferromagnetics are built up of domains is no longer an *ad hoc* hypothesis; the conclusion that there is this domain structure is reached inductively from a very wide range of experimental facts, and it is reached deductively from very general theoretical premises. Further developments of the theory, and a more complete correlation of the experimental facts, will undoubtedly be effected in the near future.

To prevent misunderstandings it should perhaps be noted that the term "domain" has been widely used in two slightly different senses. With the general interpretation adopted here, a domain may appropriately be regarded as a region of uniform strain, in which the crystal lattice approaches its ideal regularity. In such a region the magnetization will also be uniform. In the phrase "shift of domain boundaries", "domain" is used in the second sense, of a region of uniform magnetization. With this sense magnetization to saturation could be regarded as fusion into a single domain, coterminous with the specimen. There is much to be said against this second usage, in which domains assume a somewhat intangible and evanescent character; but it is frequently useful in leading to brevity of statement, the significance of which should be clear from the context.

SUMMARY AND APPLICATIONS

A few of the conclusions which have been reached about the main characteristics of magnetization curves are summarized below, with reference particularly to polycrystalline material.

(1) The saturation magnetization per unit mass, I_0/ρ, is a primary characteristic. It depends on the chemical constitution of the material, and is affected little, if at all, by physical treatments which leave this unchanged. The manner of variation of I_0 with temperature is in good general agreement with that calculated for the spontaneous magnetization on the basis of a molecular field treatment on the supposition that the ultimate elementary magnets are electron spins.

An interpretation of the molecular field is afforded by quantum mechanics.

(2) The manner of approach to saturation is determined mainly by the magnetic anisotropy of the crystalline grains. The process involved is a rotation of the direction of magnetization from "easy" axes towards the direction of the field. The natural anisotropy may be profoundly modified both by applied stresses and by internal stresses.

(3) The remanent intensity I_r ($B_r = 4\pi I_r$) is dependent on the distribution of easy axes in the crystals. Assuming random distribution of the crystal grain directions, ideally, for a single easy axis, $I_r/I_0 = 0.5$; for cubic crystals, with the cube edges as "easy" axes (as for iron), $I_r/I_0 = 0.831$; with the cube diagonals (as for nickel), $I_r/I_0 = 0.866$. With increasing tension (compression) applied to material with positive (negative) magnetostriction, I_r/I_0 approaches unity; with negative (positive) magnetostriction, zero. For a random distribution of internal pressures and tensions, I_r/I_0 will approach 0.5 as their magnitude increases.

(4) The initial increase in magnetization in low fields involves primarily a reversible shift of boundaries between regions of differently directed magnetization, domain boundaries. Consideration of a particular model indicates that the maximum value of the initial susceptibility, κ_0, is of the order $I_0^2/\pi\lambda Z_i$, where λ is the magnetostriction coefficient, Z_i a measure of the internal stress amplitude. Owing to the magnetostrictive strains set up in cooling down from the Curie point, there is a lower limit to Z_i, of the order $E\lambda$, where E is Young's modulus, this corresponding to a theoretical upper limit for κ_0, of order $I_0^2/\pi\lambda^2 E$. More detailed consideration shows that κ_0 is inversely proportional to the average stress gradient at the domain boundaries, and that only if the form of the stress distribution is known can it be related directly to the stress amplitude. Essentially it is inhomogeneities of stress within the grains which are primarily determinative of initial susceptibility.

(5) Coercivity, H_c, is a measure of the average value of the field necessary for the initiation of the irreversible, spontaneous boundary shifts with which the Barkhausen effect is associated. The critical field for any domain will be proportional to λZ_i, where Z_i is a measure of the maximum stress difference in the neighbourhood of the boundaries of the domain. Both H_c and $1/\kappa_0$ depend on inhomogeneities of stress within grains, but in different ways, H_c being a measure of average stress amplitudes, $1/\kappa_0$ of average stress gradients.

The form of the magnetization curve in low and moderate fields depends mainly on the range of magnitudes, and the distribution, of inhomogeneities of stress.

Applications. The principles of interpretation which have been outlined are of value in providing a guide through the confusing maze of empirical data relating to the magnetic properties of materials, and in suggesting lines of attack on the problem of controlling these properties, and of producing materials with properties of a desired kind. For a discussion of particular materials, which is outside the scope of this paper, on the basis of the general principles described here, reference may be made to an admirable survey by Kussmann (1935). It may be well, however, to refer briefly to one or two points as illustrative of the bearing of the discussion.

High permeability material. The essential requirement for high permeability is the reduction of internal strains. Strains of mechanical origin can be largely reduced by suitable heat treatment. Those due to the presence of "impurity" atoms depend on the manner in which they occur. In strict solid solution, the effect of such atoms is very small. (Thus the coercivity at first remains practically constant on addition of molybdenum to iron, or of copper to some of the ferronickels, but increases rapidly when the limit of solid solubility is reached.) If a second constituent segregates in relatively large masses (e.g. carbon as graphite in iron) its effect is relatively small. It is in intermediate forms that the effect is most marked, the internal strain effects then being a maximum. It is not chemical purity which is essential, but homogeneity of strain, and much of the technique of production of magnetic materials is thus successfully aimed not at the removal of impurities but in rendering them harmless. (The relatively enormous effect which even minute amounts of impurity may have is shown particularly by Yensen's work on iron.) Internal inhomogeneities of stress are less effective the smaller the magnetostriction, so that the high permeability of alloys in the permalloy region becomes at once intelligible. It has been suggested that the quenching treatment is here effective in that it results in the retention of impurity atoms in solid solution. The process of cooling in a magnetic field leads to a great enhancement of the permeability of some of the ferronickels (Bozorth and Dillinger, 1935), which is immediately attributable to the "freezing in" of the magnetostrictive strains in such directions as to give "easy" axes in the direction of the field.

Permanent magnet material. For permanent magnets, the strain in-homogeneities must be as large as possible. A quenching treatment in general gives rise to increased mechanical strains, but it is not necessarily the most effective means of obtaining permanent magnet qualities. Two or more component systems are required. Quenching may result in the production of more or less heterogeneous mixtures, in which high temperature phases may persist. In the nickel-aluminium magnet steels, neither slow cooling nor quenching is appropriate. A general procedure is quenching followed by a tempering process when precipitation may occur to a controllable extent, though short-circuiting may be possible by the choice of a suitable rate of cooling. The recent work of Bradley indicates that the best magnetic qualities correspond to a state of incipient segregation into two phases rather than the completion of the process, a result in agreement with the view that it is the state of maximum inhomogeneity of strain which is required. The theoretical indication that high magnetostriction is a favourable characteristic in materials for permanent magnets is borne out by the magnetostrictive characteristics of cobalt steels, though there are not sufficient data available in connexion with the more complex alloys.

These rather random notes on high permeability and permanent magnet materials may be sufficient to suggest the kind of problem in connexion with which the more general ideas described in this paper may be of service. The use of powder materials, particularly in high-frequency technique, involves principles which are more generally known. Such questions as those of grain size, and pre-ferential grain orientation have not been discussed, though they may clearly be brought into relation with the general scheme. The more extended application of the general principles, and their quantitative development are, however, matters for the future; in this paper the attempt has been made to give an impression of the present position in connexion with the general problems rather than with their particular exemplifications.

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Lecture V

PERMANENT MAGNETS

BY

D. A. OLIVER, M.Sc., F.Inst.P.

Permanent Magnet Association, Sheffield

Lecture V

INTRODUCTION

$\mathbf{P}$URE iron in the annealed state has a high permeability and low hysteresis loss—and is referred to as a "soft" magnetic material. If it is hardened by cold work or by the addition of alloying constituents which produce considerable internal strains, it has a relatively low permeability and high hysteresis loss, properties which are associated with a "hard" magnetic material. Permanent magnets belong to this latter class, and we are therefore concerned with materials which are in a high state of internal strain, deliberately produced and designedly kept just below the critical value at which mechanical rupture would occur.

Complicated carbide systems have been the usual means resorted to for securing internal strain, but the newer class of carbon-free alloys has secured this end by the mechanism of dispersion- or precipitation-hardening, the details of which have recently been elucidated by Dr Bradley and his co-workers from outstanding X-ray studies.

SOME PRELIMINARY CONSIDERATIONS

The modern view of ferromagnetic materials is that they are composed of discrete aggregates of iron atoms of which the magnetic spins of the electrons contributing to the external magnetic field are aligned in one direction, and that the group as a whole is not only spontaneously magnetized to saturation but that it has the property of retaining sensibly a resultant direction of magnetization, which however can be changed by the application of an external magnetic field or by the disturbance in the balance of the internal fields by mechanical strain or deformation. When all the directions of magnetization of the groups, or "domains" as they are called, are aligned, then the intensity of magnetization is a maximum and the saturation value I_s corresponding to the saturation flux density B_s has been reached and the material is said to be fully magnetized while the applied field is present. These quantities are linked by the well-known equation

$$B_s = H_s + 4\pi I_s \text{ where } I_s \to \text{constant.}$$

When the applied field is removed the flux density in a closed circuit

of the material falls to a new value at $H=0$ equal to B_r, the "re-manance". The reversed field H_c required to reduce the induction to zero is called the "coercive force". These quantities are indicated in Fig. 1, which shows the stages in magnetization and demagnetization.

Consider a closed circuit of permanent magnet material in the fully magnetized condition. The induction per unit area is B_r, but in so far that there is no working air-gap, the stored magnetic energy cannot be utilized. Imagine now a transverse slit cut in the ring. In the gap so formed a magnetic field will be established but the induction in the material will have fallen to a new point B_w on the de-magnetization curve, called the "working point". It is easy to show that the greater the reluctance of the equivalent gap associated with the magnet the lower is the point B_w on the curve, but in so far that

Fig. 1. Curves showing stages in magnetization and demagnetization.

the curve is usually plotted in units whereby the flux density B is quoted in gauss and the magnetizing force H in oersted, the other important variable associated with a given working point on the demagnetization curve is the ratio of length L, to cross-section A, of the magnet part itself, and this is often termed the "dimension ratio". The simplest relation connecting the reluctance R and the dimension ratio of the magnet (L/A) is

$$R = \tan \psi (L/A),$$

where $$\psi = \tan^{-1} (H_w/B_w).$$

The energy which a given volume of magnet steel can set up in the working gap of a given external magnetic circuit depends on the particular shape of the demagnetization curve for the material and on the actual working point on that curve, which can be controlled by the dimension ratio chosen for the magnet part itself. Elementary theory shows that the energy associated with any point taken on the demagnetization curve depends upon the product of B and H, and this is a maximum for a point lying approximately midway between the remanence and coercive force terminal values. A simple graphical

method of deciding the position of this point has been given by Watson and is amply accurate enough for all practical purposes (Fig. 2).

To achieve the optimum design whereby the magnetic energy set up in the working gap is secured by a minimum volume of material, it is necessary to arrange that the working point of the completed magnet system coincides with the BH_{max} point on the demagnetization curve. If this condition is satisfied then for a given working gap in the magnetic circuit there results one particular dimension ratio for the magnet part. Different dimension ratios can, however, be used in practice provided a lowering in magnetic utilization efficiency can be tolerated. In certain cases other factors influence the design and make it necessary to depart from the dimension ratio determined from minimal material considerations. An example of this is found in the well-known case of electricity meter magnets made from the less expensive steels, where an excessive length ensures minimal changes from external demagnetizing influences and the factor of permanence is more important than slight economy in steel. An

Fig. 2. Demagnetization curves for British "Alnico" showing Watson's BH_{max} construction.

illustration of the reverse state of affairs where the length is made a minimum and the section a maximum is found in certain loud-speaker magnets where back to front depth has to be kept small. As a general rule, if the total volume of magnet material used is not a critical factor, it is greatly to be preferred to have an excess of length rather than an excess in section, or in other words a dimension ratio greater than that determined from optimum design considerations.

The most important single criterion of a permanent magnet alloy is the product BH at the maximum point. This value divided by 8π gives the theoretical optimum magnetic energy in ergs per c.c. of material which can be set up in any external magnetic circuit associated with it, and in what follows will be regarded as the primary criterion. Other criteria have in time past been put forward and used extensively and the most important of these has been the product $B_r H_c$, but this is only valid because the general shape of the de-

magnetization curve between these two points is approximately constant and because

$$BH_{\max} = K\,(B_r H_c),$$

where K is a constant and is termed the "fullness factor". Its value is approximately 0·42 for a number of materials. This relationship is popular because B_r and H_c are easily observed whereas the value of $BH_{\max}$ requires greater experimental elaboration, but instances could be cited where the product $B_r H_c$ is not only misleading but is insensitive to improvements made in the material by careful heat-treatment. This again is a further reason for insisting on an experimentally determined value of $BH_{\max}$ as the only reliable standard by which to appraise the true magnetic efficiency of a material. Different applications demand the desirability of different-shaped demagnetization curves, but the normal procedure is to develop a material giving the greatest value of $BH_{\max}$ and then to design the permanent magnet system employing it with due regard to the desirable dimension ratio.

MAGNETIZING AND DEMAGNETIZING

Many visitors to the works of Messrs William Jessop & Sons Ltd. of Sheffield, with whom the author is associated, when inspecting the magnet production department have asked straight away to be shown how the steel is magnetized. Some have expressed surprise that even when the casting or forging, annealing, machining, heat-treatment, and hardening operations are all carried out the magnet still exhibits no external magnetic effects. This is explained by the fact that the domains in all these materials are not only spontaneously magnetized but that the directions of magnetization are arranged at random, satisfying a condition of minimum energy associated with no external magnetic effects. The final process of magnetization can be likened simply to the alignment of the magnetizations of the domains, which are then prevented from swinging back into a random arrangement by the already skilfully placed atomic arrangements surrounding the domains and acting as constraints preventing their easy rotation. The greater the proportion by volume of the domains the higher is the value of the remanence B_r; the more efficient the constraining action, the greater is the value of the coercive force H_c. To continue this conception a stage further in an ordinary carbon steel a relatively large proportion of the material is filled by the domains, but they are

subject to poor constraints. In the nickel-aluminium-iron alloys on the other hand the total volume of the domains is lower but the atomic constraining arrangements are very efficient.

Let us consider for a moment the magnetization of a simple bar magnet. If this is placed in the axis of a solenoid having a high value of axial field the bar magnet can be completely saturated, but in practice for certain materials the value of the field has to be so high that this method is not practicable. As a method it implies that the ampere-turns per unit length of the coil have to be very high and furthermore because the bar is on open-circuit an excess field has to be provided so that the applied field less the self-demagnetizing field of the bar magnet is equal to the field required for saturation. An obvious development improving the magnetizing conditions is to close the bar magnet by a soft iron yoke system, and when the coil has been wound on the iron yoke and adjustments have been provided to vary the gap so that different lengths of bar can be accommodated, we have a well-known form of magnetizing arrangement which is usually referred to as a "contact magnetizer".

There is one form of magnetizing arrangement which does lend itself to the production of a high value of magnetizing field where the ampere-turns per unit length of the coil are high. In this case the turns are a minimum, possibly one or two turns of a stout copper bar, and the current is momentarily very high indeed, of the order of 10,000–20,000 amp. This current is obtained by supplying the primary of a transformer with direct current and short-circuiting a single secondary turn by means of the one or two loops of massive copper already mentioned. On breaking the primary circuit the flux in the core of the transformer collapses and a very heavy current is generated for a short time in the low-resistance secondary circuit of which the copper loops form a part. In order to get the best effect, the iron circuit of the transformer has a large series air-gap and this enables the flux to collapse quickly thereby generating a higher value of peak short-circuit current. To saturate the newer permanent magnet alloys a minimum peak magnetizing force of approximately 3000 oersted is required and about twice this value is really to be preferred.

For demagnetizing it is desirable to have slowly varying cycles of magnetization which start initially with a peak value almost sufficient to magnetize fully in the ordinary way. Providing the field does not vary too rapidly the demagnetization takes place uniformly through-

out the section of the permanent magnet material and this is especially important with the modern high coercive force alloys. The problem is a real one and even now experiments are being conducted to find a really efficient method of carrying out the operation. Reversed magnetization of the correct amount is satisfactory but in all but special instances the method is impracticable on account of the precise control demanded.

Small magnets can be demagnetized by placing them within a multilayer coil carrying a large alternating current at a frequency of 50 c.p.s., but for magnets of considerable size and section, even if the peak field can be established, the frequency proves too high and the demagnetization is not perfect.

BRIEF HISTORY OF PERMANENT MAGNET STEELS AND ALLOYS

Before 1910 the principal permanent magnet material was glass-hard carbon steel containing approximately 1–1·5 per cent of carbon, and large numbers of carbon steel magnets were used where cheapness was the first consideration. These magnets were very susceptible to temperature variations and to mechanical shock and vibration and were only successful where excessive dimension ratios in design were employed.

The origins of tungsten magnet steel are wrapped in mystery, but a metallurgist Willis knew of tungsten magnet steel about 1880, although Remy and Bohler claimed to have invented it on the continent about 1882. However, in 1913 Silvanus Thompson reported that there were two types of tungsten steel in common use, one having 0·5 per cent carbon and 5·5–8 per cent tungsten while another variety contained 1·0 per cent carbon and 2·5–3·5 per cent tungsten. The first composition was said to give better results but the latter was used sometimes because it was less expensive. A good modern tungsten magnet steel contains 0·67 per cent carbon, 0·35 per cent chromium and approximately 6 per cent tungsten, and it is clear therefore that the first composition mentioned by Silvanus Thompson was very much nearer the optimum composition than the second one. The addition of chromium appears to be of comparatively recent date, because the classic investigation of Evershed in 1920 on tungsten magnet steel does not specifically deal with the effect of chromium and there is reason to believe that he used only chromium-free steels. Prior to Evershed's research Madame Curie investigated the magnetic

properties of tungsten steel and concluded that a carbon content of
0·6 per cent associated with a tungsten content of 5–6 per cent was
about the best composition and this result is certainly not very far
from the optimum composition already given.

In 1909 Prof. Brown in Dublin investigated chromium steels from
the point of view of their permanent magnetic properties and
concluded that a steel containing 0·86 per cent carbon and 1·95 per
cent chromium gave the best results and that no advantage accrued
from using a chromium content exceeding 3 per cent. Dr Matthews
in America claimed that chromium magnet steels resulted from
routine observations of alloy steels and that the first oil-hardening
chromium magnet steel was really a commercial chromium hot-work
tool steel, but it is fairly certain that the first systematic work was
carried out by Prof. Brown in 1909. During the last war Germany
was short of supplies of tungsten and consequently fresh interest was
shown in the use of chromium steels for magnets, and Gumlich in
1917 put forward a 6 per cent chromium steel which has now secured
a prominent place among the modern steels. A typical composition
is 1·0–1·1 per cent carbon and 6 per cent chromium, and such a steel
suitably heat treated will give results almost equal to 6 per cent
tungsten steel with the exception that the remanence value may be
a little less, but in energy content and coercive force there need be
nothing to choose between them.

In 1917 Honda and Takei discovered that the addition of cobalt
and tungsten to a carbon steel produced an astounding improvement
in the magnetic properties, and the outcome of this work was the
famous 35 per cent cobalt steel which is still in widespread
use. The modern 35 per cent cobalt steel has a somewhat greater
carbon content than those put forward in 1920 when the results
were published, but in all essential respects it is practically the same.
In· the hands of many investigators intermediate steels containing
smaller percentages of cobalt and larger percentages of chromium
soon appeared, and to-day there is a recognized range of cobalt
steels containing 3, 6, 9, 15 and 35 per cent cobalt, the steels with the
lower cobalt contents having greater chromium contents.

The steels already described held the field until about 1934, when
as a result of a remarkable discovery made by Prof. Mishima in
Japan in 1931 the whole outlook on permanent magnet materials
was changed. He was experimenting with ternary alloys of nickel,
iron and a third element, and when this element was aluminium, for

certain compositions, he found that his test-pieces showed abnormally high mechanical hardness and brittleness. Because his investigations included an approximate examination of magnetic properties he noticed that these alloys possessed exceptionally large values of coercive force and a new chapter commenced in the history of permanent magnet materials. It is interesting to look back and realize now that ternary alloys of iron, nickel and aluminium had been made in this country and used as a cutting-tool material but their extraordinary magnetic properties had never been suspected until Prof. Mishima made his investigations. The optimum magnetic properties are associated with a composition of the order of nickel 29 per cent, aluminium 13·5 per cent, with iron the remainder, or practically with a composition corresponding to Fe_2NiAl. About the same time as Mishima's discovery, Rogers and Seljesater in America and Köster in Germany found that carbon-free alloys could have outstanding properties from a permanent magnet standpoint. It was found that a ternary alloy of iron and cobalt with either tungsten or molybdenum had valuable magnetic properties for a composition of approximately iron 67 per cent, cobalt 15 per cent and tungsten 18 per cent. These particular alloys, however, have not come into use commercially on account of their high cost.

A further important discovery was made about 1933 by Honda, Masumoto and Shirakawa when they found that a ferrous alloy of cobalt, nickel and titanium could have exceptionally good properties as a permanent magnet, and indeed this material is the best from a performance point of view of any known, but is not yet available commercially.

In 1934 Horsburgh and Tetley in Sheffield studied the effects of the addition of cobalt to the ternary nickel-aluminium-iron system. Mishima had previously mentioned cobalt as an improving element and its presence is undoubtedly beneficial. However, it was found difficult to obtain good magnetic properties except in very thin cast sections, but Horsburgh and Tetley overcame these difficulties by finding that the addition of copper was essential to achieve the best results. By systematic experiment the best composition was arrived at and as a result of this work the modern material known in this country as British "Alnico" resulted, which is commercially available. Its composition is approximately nickel 18 per cent, aluminium 10 per cent, cobalt 12 per cent, copper 6 per cent and iron 54 per cent, and is undoubtedly superior to any composition

containing cobalt without the presence of copper. In Japan, Mishima's improved alloy, copper-free but containing cobalt, is called "M.K. steel", and Honda's new alloy containing titanium is called the "New K.S. steel", but the author is strongly adverse to calling this class of alloy a steel because carbon is absent and there seems little justification, if any, for the use of the word "steel".

In 1935 another interesting carbon-free ternary alloy of nickel, copper and iron was announced by v. Auwers and Neumann in Germany. This alloy, which for optimum properties contains 20 per cent nickel, 60 per cent copper and 20 per cent iron, is distinguished by two major features. The first is that it can be forged, drilled, tapped and easily machined, and the second is that the magnetic properties from a permanent magnet standpoint can be greatly enhanced by lattice distortion due to cold-work. It has been given the name "Magnetoflex" in Germany.

A short review of the history of these magnetic materials would not be complete without a reference to the very original work of Kato and Takei who have mixed the oxides of iron and ferro-cobalt, Fe_3O_4 and $Co.Fe_2O_4$, which are prepared by an electrochemical method. Equal quantities of the oxides are taken, mixed intimately together and compressed into the desired form of magnet, which is then sintered at 1000° c. and magnetized at 300° c. to secure the best results. The density of the resulting material is only 3·55 g./c.c.

THE MAGNETIC PROPERTIES OF THE AVAILABLE MATERIALS

Having already defined the magnetic energy criterion BH_{max}, the remanence B_r, and the coercive force H_c, it is now easy to show these properties in diagrammatic form to facilitate their intercomparison. In Figs. 3, 4, and 5 are shown the BH_{max} values, the B_r and the H_c values respectively. To portray the properties in this way is of value from a design standpoint because some applications naturally demand a material with high remanence while others require a high coercive force. In other instances the major consideration is over-all size and this therefore requires a material of high intrinsic magnetic energy.

The magnetic saturation intensity and the Curie temperatures for these ferromagnetic steels and alloys are of great theoretical interest. As yet, however, very few systematic measurements are available but a few representative ones are as follows: pure iron has an intensity of
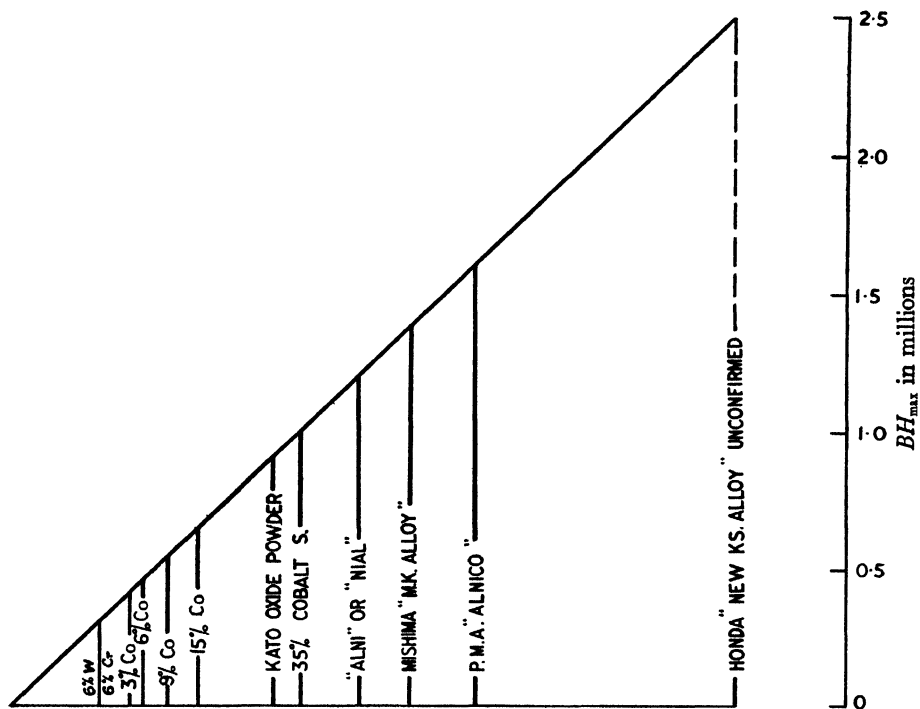
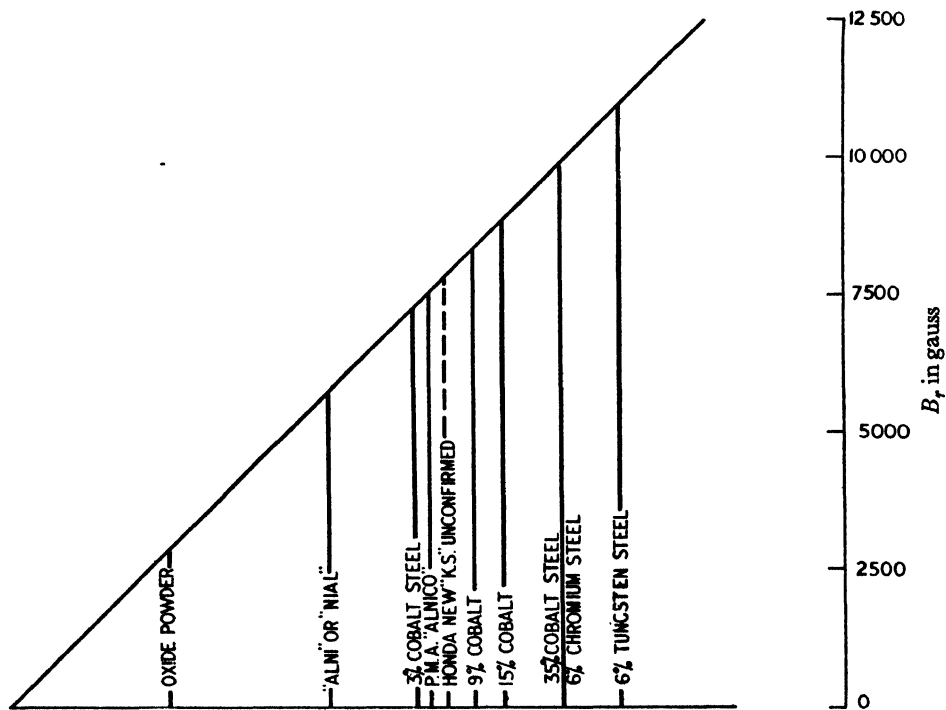Fig. 3. Magnetic energy of permanent magnet materials.

Fig. 4. Remanence of permanent magnet materials.

magnetization I_s equal to 1720 while 35 per cent cobalt steel has a value of I_s equal to 1300, and a copper-free Mishima alloy containing cobalt has a value I_s equal to 815.

The Curie temperatures for 6 per cent tungsten, 35 per cent cobalt and the new nickel-aluminium-iron alloys are all in the neighbour-
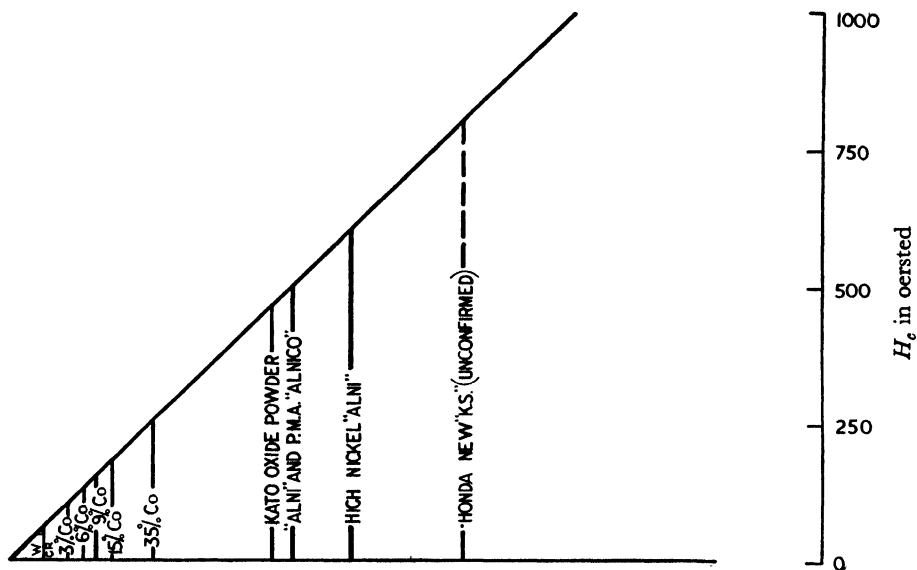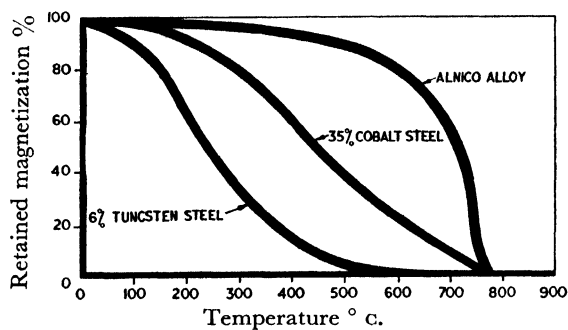
Fig. 5. Coercive force of permanent magnet materials.

Fig. 6. Curves showing relative fall-off in magnetization with temperature.

hood of 710–760° c., which is the temperature for the ordinary magnetic change point for iron (766° c.). Nevertheless in Fig. 6 the forms of the curves delineating the fall-off in magnetization with temperature are vastly different and explain why the newer alloys are superior in permanence at temperatures up to a few hundred degrees Centigrade.

In a number of practical applications the ability of a permanent magnet alloy to retain its permanence under the influence of alternating magnetic fields is important. A good example of the industrial importance of this property is afforded by the electricity energy meter industry where even 1 per cent change in the strength of the

Fig. 7. Curves showing relative fall-off in magnetization with alternating demagnetizing field.

braking magnets due to external alternating stray fields would be very serious. Further, instruments mounted on switchboard panels often have bus-bars passing near them carrying heavy alternating currents and stray fields are sometimes unavoidable. Without going into detail it is interesting to obtain an approximate idea of how the different materials vary in their ability to withstand a given peak value of alternating magnetic force. Fig. 7 shows a comparison between chromium and tungsten steels, 35 per cent cobalt steel, and Alnico which is representative of the newer alloys. Fig. 8 shows a comparison between the new oxide magnet of Kato and Takei and

Fig. 8. Comparison between oxide powder material and carbon steel in A.C. field.

quenched carbon steel. It is true that such a comparison is a little unfair in so far as the permanence of carbon steels under alternating forces is notoriously poor, but it serves to illustrate the very large differences in properties between different materials. The measured demagnetization curve for some samples of Kato and Takei's oxide
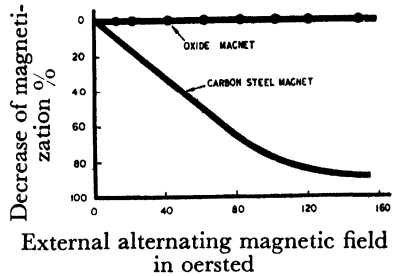
powder bars is given in Fig. 9, and the start of the curve for British Alnico is also given for comparison purposes. Due to the unusual form of the curve the *BH* product is also plotted and the BH_{max} point ascertained. There is no doubt that these oxide magnets represent an entirely new approach to the permanent magnet problem. The curves claimed by the Japanese workers are shown in Fig. 10.
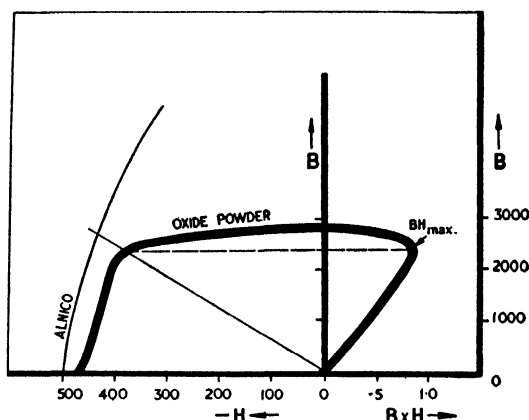

Fig. 9. Measured demagnetization curve for some of Kato and Takei's oxide powder bar magnets.

It has become customary in the literature on permanent magnets to quote comparative mechanical drop tests for specified sizes of bar and while these undoubtedly throw some light on the magnetic permanence of a material undergoing mechanical shock the results are rarely strictly comparable. This arises because if the same size of test piece is retained (that is a constant dimension ratio), the working points on the demagnetization curves for materials of widely different properties are far from comparable and the author considers that in such tests it is more important to keep the reluctance line a constant than it is to retain the same value of dimension ratio. However, some representative orthodox drop test results are shown in Fig. 11.
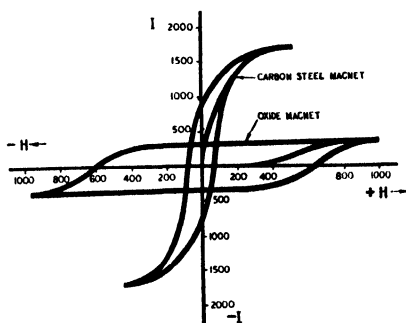

Fig. 10. Hysteresis loops of oxide powder material according to Kato and Takei.

MECHANICAL PROPERTIES

Carbon, chromium, tungsten and cobalt steels can all be worked hot and rolled into bar and subsequently hot-forged to shape in suitable tools. They are mostly hard steels but can be machined and drilled after suitable annealing, and ground to finished dimensions after heat-treatment and hardening.
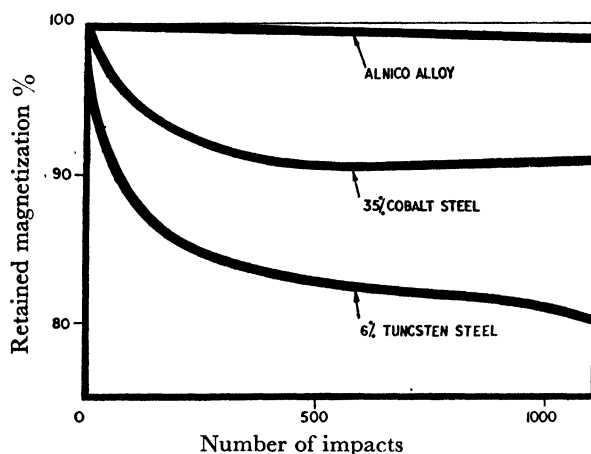

Fig. 11. Curves showing relative fall-off in magnetization with mechanical impact.

The new alloys of nickel, iron and aluminium with other alloying elements are dispersion-hardening alloys, cannot be worked hot, are very hard and brittle in the cast state, and with few exceptions cannot be appreciably annealed, but with difficulty some compositions can be turned and drilled. The normal procedure is to cast to size and finish by grinding, usually after heat-treatment. The oxide powder magnets are also very brittle.

PRINCIPAL MARKETS FOR PERMANENT MAGNETS

The principal industrial fields calling for finished magnets or magnet parts are the radio industry, and the instrument-making industry in its broadest sense.

The radio industry. It is not widely realized that the annual consumption in this country of complete magnets and magnet parts for permanent magnet moving-coil loud-speakers is in the neighbourhood of one million. This field of application is one of great importance and at present is satisfied by cast magnets in Alni and Alnico. The newer alloys have been adopted universally because high magnetic energy

content is desired and the magnet part lends itself to efficient design. A typical modern ring-type magnet is shown in Fig. 12.

The instrument-making industry. This broad classification can be subdivided with increased clarity into the following specialized industries: (1) electricity energy meters; (2) telephone and communication apparatus; (3) indicating instruments; (4) magnetos, small
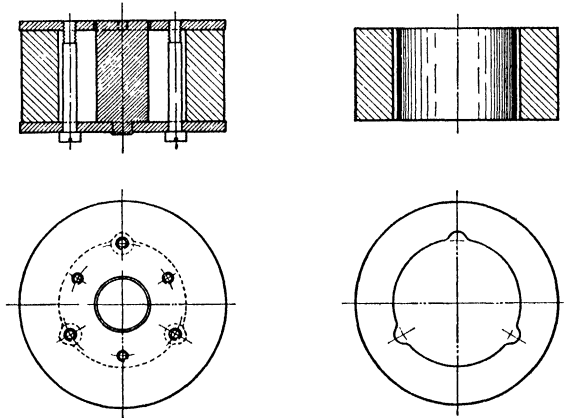

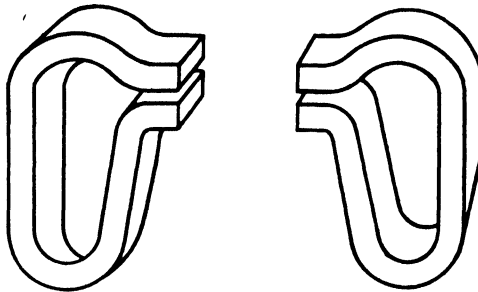Fig. 12. Typical modern ring-type moving-coil loud-speaker magnet showing ring separately.

Fig. 13. Typical forged steel electricity meter magnets.

motors and generators; (5) scientific apparatus and miscellaneous applications.

In Fig. 13 is shown a typical pair of forged steel magnets in 6 per cent tungsten or 6 per cent chromium steel such as are extensively employed to damp by eddy currents the rotating discs in electricity energy meters. Many millions of such magnets are produced annually, but in recent designs of meters, cast Alnico

magnets such as are shown in Fig. 14 are in some instances being adopted. The magnets employed in telephone receivers are of simple form. They are either small rectangular bars of 35 per cent cobalt steel ground on one face, or small cast blocks of the newer alloys ground on opposite parallel faces and clamped between soft iron components.

Indicating instruments such as ammeters and voltmeters, especially of the switchboard mounting type, have usually been fitted with simple forged horseshoe-shaped magnets, ground to size in the gap.
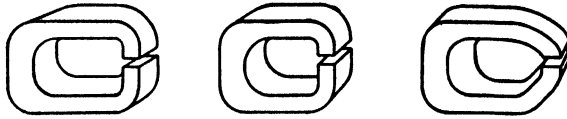
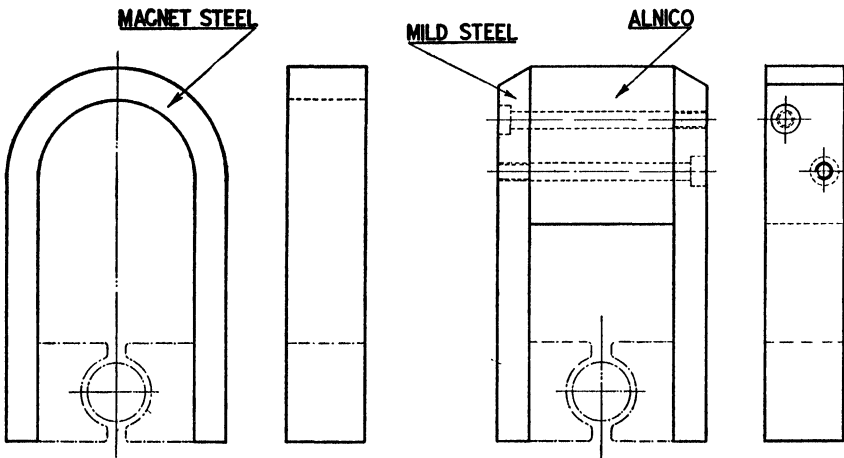Fig. 14. Examples of modern cast Alnico electricity meter magnets.

Fig. 15. Forged steel and cast alloy horseshoe-type instrument magnets.

The modern counterpart is a block of alloy clamped between mild steel limbs. Both these patterns are illustrated in Fig. 15, and in Fig. 16 are seen the corresponding magnets representative of the miniature type of instruments. The low value of remanence of the newer alloys demands in design a large area of cross-section in the magnet part and the flux is collected by the mild steel limbs over this area.

Magnetos, small motors and generators either employ magnets of the type shown in Fig. 15 or are fitted with magnet components specially shaped to suit the particular design.

CONCLUSION

In concluding it is interesting to reflect that two determinative factors have resulted in the outstanding success of the Japanese in the field of permanent magnet investigation. The first was that the late Sir James A. Ewing, at the time when he was studying magnetic phenomena, spent about two years in Japan as Professor of Physics and Engineering, and his inspiring teaching had a fundamental and lasting effect on those who were fortunate in being his students. The other powerful factor was the return of Professor Honda, at that time a research student, from Göttingen, where he was first stimulated to study metals and their magnetic properties. If we in this country
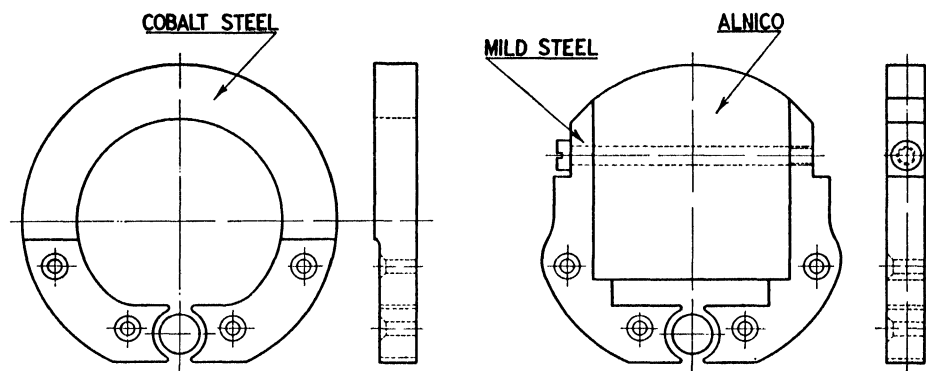

Fig. 16. Cast steel and cast alloy magnets for miniature instruments.

have been slow in the past in carrying out pioneer work on new alloys, one answer is that the recent interest in the physics of metals augurs well for the future.

Finally, it is well to remember that this country produced the great father and pioneer of all permanent magnet investigators, the great Elizabethan Gilbert, whose book *De Magnete* (1600) will ever take the first place in the literature of the subject, and doubtless we can all join with Dryden when he wrote on the occasion of the first centenary after Gilbert's death, "Gilbert shall live till loadstones cease to draw".

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Lecture VI

X-RAY STUDIES ON PERMANENT MAGNETS
OF IRON, NICKEL AND ALUMINIUM

BY

A. J. BRADLEY, D.Sc.

Royal Society Warren Research Fellow, National Physical Laboratory

AND A. TAYLOR, Ph.D.

University of Manchester

Lecture VI

INTRODUCTION

PROMINENT among the new materials[1] used for permanent magnets are the ternary alloys of iron, nickel and aluminium of the composition Fe_2NiAl. It appeared probable that these especially good magnetic properties were associated with the atomic structure of the ternary alloy, and an exhaustive X-ray investigation was carried out, on behalf of the Permanent Magnet Association, to test this hypothesis. The results of the investigation, which are of considerable interest, are given in this paper.

The X-ray work included the examination of the binary systems iron-nickel, nickel-aluminium, iron-aluminium, and the relevant part of the ternary system iron-nickel-aluminium. The materials were examined in the form of powder suitably heat treated, the technique in general being similar to that described elsewhere[2].

The first object of our investigation was to find what type of structure, face-centred cubic or body-centred cubic, was present in the alloy after slow-cooling. The alloy may contain only one type of lattice or it may be a mixture. In some cases where there are two phases, both have the same type of lattice, either both face-centred cubic or both body-centred cubic. In such cases the two phases are distinguished by differences in lattice spacing. Most of the lines of the two X-ray patterns then coincide, but those which are most resolved usually show up the difference.

In some instances where there are two lattices of the same type present on the X-ray photograph it happens that one of the two has a superlattice. That is, the corners of the cube differ from the centres of a cube, if the structure is body-centred cubic. This may provide an additional means for distinguishing two phases of the same lattice type.

THE NICKEL-ALUMINIUM SYSTEM[2,3] (Fig. 1)

Pure nickel is face-centred cubic. On adding aluminium, we obtain a solid solution in which the aluminium atoms replace the nickel atoms at random. Over 10 per cent of the nickel atoms may be so replaced. Beyond this point the solubility depends on the heat treatment, until we reach almost 25 atomic per cent of aluminium.

At this composition the face-centred cubic lattice forms a single phase, and the atoms take up an ordered arrangement. The nickel atoms go to the centres of cube faces, the aluminium atoms to cube corners.

X-ray powder photographs of alloys containing between 10 and 25 atomic per cent of aluminium show some very interesting effects. They all have face-centred cubic patterns, but with different lattice
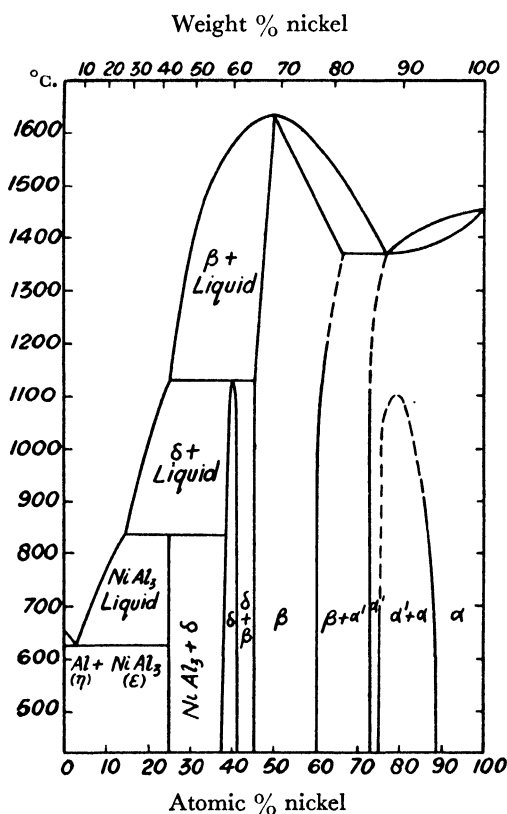

Fig. 1. The nickel-aluminium system. *Reproduced by permission from the Proceedings of the Royal Society.*

spacings. The greater the aluminium content, the higher the spacing. On slow cooling, they break up into two constituents, one with a high spacing, the other with a low spacing. The former corresponds closely to Ni_3Al, the latter contains much more nickel.

After quenching the powders from 700° c., the lines of the two patterns come much closer together, showing that the range of limited miscibility is contracting. This is in accordance with the phase equilibrium diagram (Fig. 1).

When more than 25 atomic per cent of aluminium is present in the binary alloy with nickel the face-centred cubic structure soon breaks down. At 40 atomic per cent of aluminium, the structure becomes body-centred cubic, and alloys containing between 25 and 40 per cent aluminium are mixtures of two phases, one face-centred cubic, the other body-centred cubic.

In the body-centred cubic phase of the nickel-aluminium system there is a regular arrangement of the atoms, aluminium and nickel sorting out as far as possible between cube centres and cube corners. Ideally, the ordering process can only be complete at the exact composition, NiAl. With less aluminium, nickel is able to replace aluminium up to the phase boundary. The alloy may be regarded as a solid solution of nickel in NiAl.

THE IRON-ALUMINIUM SYSTEM [4, 5, 6]

The iron-aluminium system bears some analogy to the nickel-aluminium system. There is the same difference between the aluminium half of the system and the rest. With less than 50 atomic per cent aluminium, there are only two kinds of structure. Just as in the nickel-aluminium system, these are face-centred cubic and body-centred cubic, respectively. FeAl is exactly like NiAl; the structure is body-centred cubic and there is a complete sorting out of iron and aluminium atoms between cube centres and cube corners. On the phase diagram (Fig. 2) we have illustrated this analogy by calling the phase by the same Greek letter as NiAl (β), although this is contrary to general usage.

The large area of body-centred cubic structures on this diagram has been split up into a number of "subphases". These differ in atomic arrangement and in their magnetic properties. Near pure iron there is no regular ordering of the iron and aluminium atoms. Near Fe_3Al there is a special type of ordered structure, where one position in four is reserved for aluminium. As the aluminium content increases, this structure changes to the FeAl type. Suffixes are used to distinguish the three types of body-centred cubic lattice, and to show which portions of the diagram include magnetic alloys. These are marked m, the non-magnetic alloys being n. The two magnetic change-points in Fe_3Al were found by Sykes and Evans [6].

THE IRON-NICKEL SYSTEM [7, 8, 9, 10]

The iron-nickel system again contains only face-centred cubic and body-centred cubic structures. In recent years it has been the subject of much study, and the position is now much clearer than a few years ago. The general result of these investigations is to confirm the correctness of a diagram such as that proposed by Merica [7], though the exact position of the boundaries is still in some doubt.
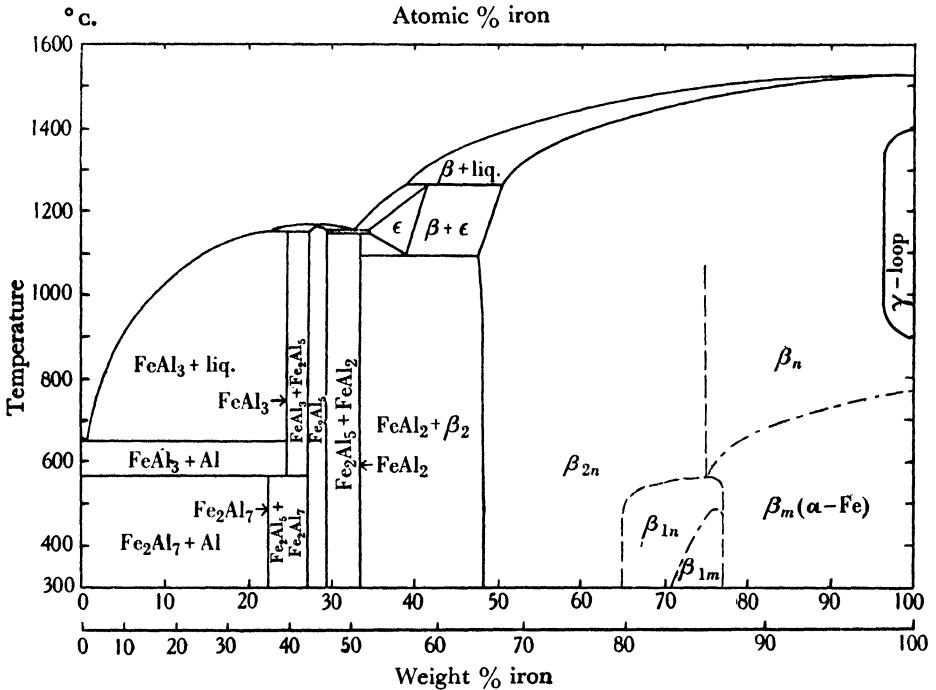

Fig. 2. Modified Fe-Al diagram.

Below 4·4 per cent nickel there is no trouble; the alloys are face-centred cubic at high temperatures and change to body-centred cubic on cooling. Beyond about 29 per cent nickel they are face-centred cubic even after cooling to room temperature. Between 4·4 and 29 per cent of nickel there is a region where the transformation of face-centred to body-centred cubic structures begins readily on cooling, but is difficult to complete. Unless very special precautions are taken, alloys cooled down to room temperature contain a mixture of the two forms. It is simplest to regard this as a two-phase region.

In ternary alloys containing iron and nickel, slow cooling to room

temperature gives results which are consistent with the above interpretation of the diagram: i.e. they show a body-centred cubic pattern from pure iron up to about 4·4 per cent of nickel, followed by a two-phase region which stretches towards 29 per cent of nickel.

It has been suggested that the iron-nickel alloys with compositions near Ni_3Fe assume an ordered structure on slow cooling. This is not borne out by our experiments on slow-cooled alloys. By the use of cobalt radiation, the scattering power of iron is depressed so much more than that of nickel that it should be possible to detect the presence of a superlattice. Jones and Sykes[11] have recently found the superlattice in CuZn, by a suitable choice of radiation. In the present instance, we can detect the superlattice when a small amount of aluminium is added to Ni_3Fe, but it is absent from pure Ni_3Fe.

THE TERNARY DIAGRAM OF IRON, NICKEL AND ALUMINIUM [12]

The pioneer work on this system was carried out by Köster, who examined the ternary alloys by the microscope and made magnetic measurements on them. He concluded that the diagram was a simple modification of the iron-nickel diagram, with face-centred cubic, body-centred cubic and two-phase areas. This is far too simple an interpretation, as we have shown by recent X-ray photographs. For example, recent work proves the existence of the miscibility gap in the nickel-aluminium α-phase. There is however a further complication in the ternary system.

Our new X-ray work shows that Köster's two-phase region is in reality a collection of four separate areas. This is possible because the body-centred cubic phase, which includes pure iron, has a most remarkable shape. In general outline, the boundary of the body-centred cubic phase follows the course shown in Fig. 3. It is clear from this diagram that the main bulk of the phase is far from the iron-nickel binary system, and can in no way be regarded as a modification of this system. Only the narrow tongue stretching along the iron-aluminium side of the diagram really corresponds to the body-centred cubic phase of the iron-nickel system.

The distinction between the iron-rich portion of the body-centred cubic phase and the rest clearly follows from the iron-aluminium diagram, as given in Fig. 2. The narrow tongue in the ternary diagram has a random distribution of atoms, like the iron-rich iron-aluminium alloys. The broad portion of the body-centred cubic

phase has the FeAl type of superlattice, in which there is a clear distinction between cube centres and cube corners. One position is as far as possible filled by aluminium, the other contains both iron and nickel.

To distinguish the ordered structure from the random, the same nomenclature is used as in Fig. 2, which we have already described. β alone indicates the random structure; β_1 is the small portion of the
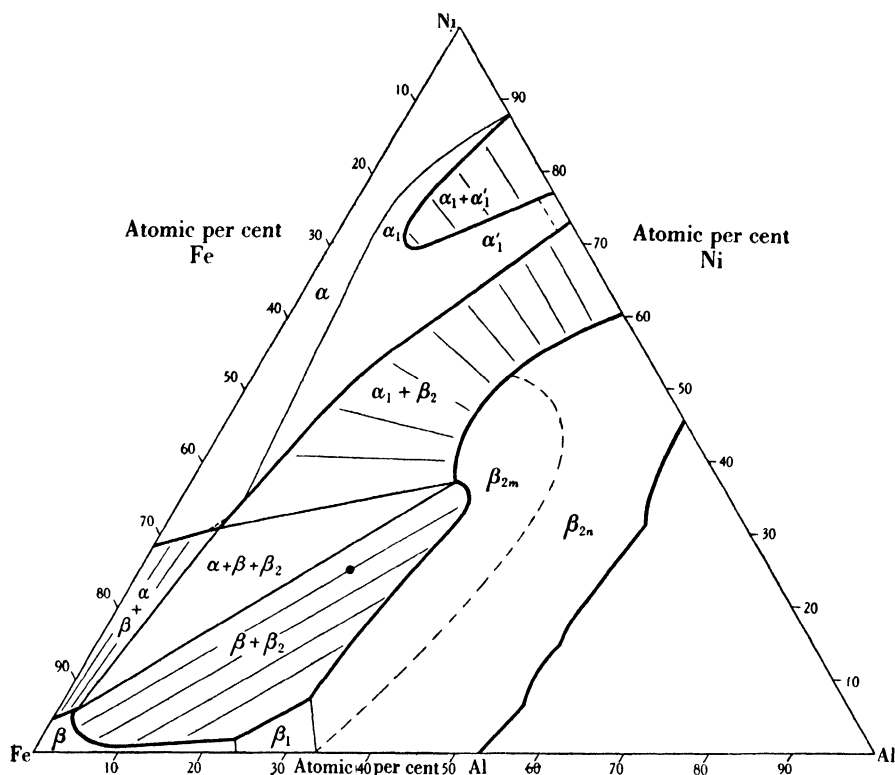

Fig. 3. Ternary Fe-Ni-Al diagram.

body-centred cubic phase with a superlattice like Fe_3Al; β_2 has a superlattice like FeAl. In the ternary diagram, the β_1 region which at first separates β from β_2 is eventually replaced by a two-phase $\beta + \beta_2$ region. The β_2 area is further subdivided into a magnetic portion β_{2m}, and a non-magnetic portion β_{2n}.

The two-phase area $\beta + \beta_{2m}$ includes all the compositions which are suitable for the manufacture of permanent magnets. Fe_2NiAl, which is indicated on the diagram, is typical. This two-phase area is shown covered by a grid of tie-lines. An alloy lying on any such tie-line

splits up into two constituents, the compositions of which are given by the ends of the lines. For example, Fe_2NiAl, which is single phase at high temperatures, breaks up into $Fe_{38}NiAl$, near pure iron, and $Fe_6Ni_7Al_7$, near the centre of the diagram, on slow cooling.

Köster had already shown that the boundary of the β-phase was very susceptible to heat treatment, and we have confirmed this by a series of quenching experiments. We find that the manner in which Fe_2NiAl decomposes on cooling can be greatly influenced by varying the quenching temperature. According to the older view, it could only break up into a mixture of the face-centred cubic and body-centred cubic phases. The new diagram (Fig. 3) shows that this is incorrect. There is none of the face-centred cubic phase in Fe_2NiAl after slow cooling; the decomposition consists solely in splitting off an iron-rich body-centred cubic constituent β from the main portion of the structure (β_2).

THE CRYSTAL STRUCTURE OF THE PERMANENT MAGNETIC STATE [13, 14, 15]

The new diagram removes the difficulties found by earlier X-ray workers on the magnetic alloys (Werestchiagan and Kurdjumov[13], Glocker, Pfister and Wiest[14], and Burgers and Snoek[15]). These authors had shown that the magnetic alloys give no face-centred cubic pattern except after quenching from high temperatures. At lower temperatures, the body-centred cubic pattern was unchanged except for a broadening of the lines. There was no appreciable difference in the lattice spacing.

Burgers and Snoek investigated the effect on the magnetic properties of cooling at different rates from 1200 to 700° c. The results are given in Fig. 4. This shows the coercive force (H_c) of a nickel-iron single crystal in relation to the time of cooling. The rapidly cooled material has a very low coercivity, and is therefore useless as a permanent magnet. A very slowly cooled specimen gives only moderately high values of the coercive force. The best results are obtained by cooling at a definite rate.

X-ray powder photographs were taken with a view to explaining these facts. Burgers and Snoek's data are given in Fig. 5. The photographs show the 310 reflection from a single crystal. The state of maximum coercivity corresponds to (e) where the line has begun to broaden. The two $K\alpha$ components ($K\alpha_1$ and $K\alpha_2$) are still visible

on the photometer record, showing that the original crystals are still present, without a great change in structure.

We have carried these experiments a stage further. In Fig. 6 we show data from the same reflection (310), but from a powder instead of a single crystal, and after different heat-treatments. Fe_2NiAl was quenched successively from 900, 800, 700, and 600° c., and finally it was slow cooled. The powder photographs show a gradual broadening of the lines as the quenching temperature is reduced, culminating in a definite doubling of each line, in the slowly cooled state.
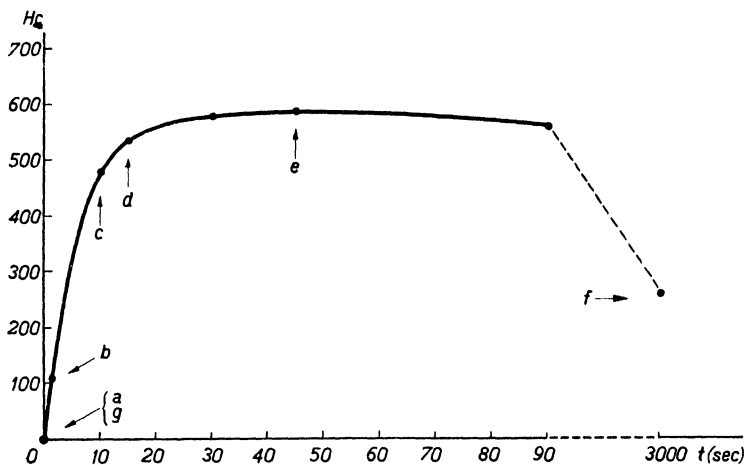

Fig. 4. Coercive force (H_c) of a nickel-iron-aluminium single crystal in relation to the time t, required for cooling from 1200 to 700° c. *Reproduced by permission from Physica.*

The doubling of the lines can be understood from the phase diagram given in Fig. 3. The slowly cooled alloy consists of two constituents of slightly different lattice spacing. The X-ray patterns of the two constituents almost, but not quite, overlap. There are therefore four lines belonging to the 310 reflection instead of the usual α doublet. One pair of lines belongs to the pattern of the iron-rich constituent, which has a lattice spacing slightly smaller than the rest of the alloy. The other pair belongs to a constituent much richer in iron and aluminium. The superlattice line which is present on the photograph also belongs to this constituent, and not to the iron-rich one.

Combining the evidence obtained by Burgers and Snoek with that from our own experiments, it is possible to picture the process which leads to the formation of the "hard" magnetic alloy. In the
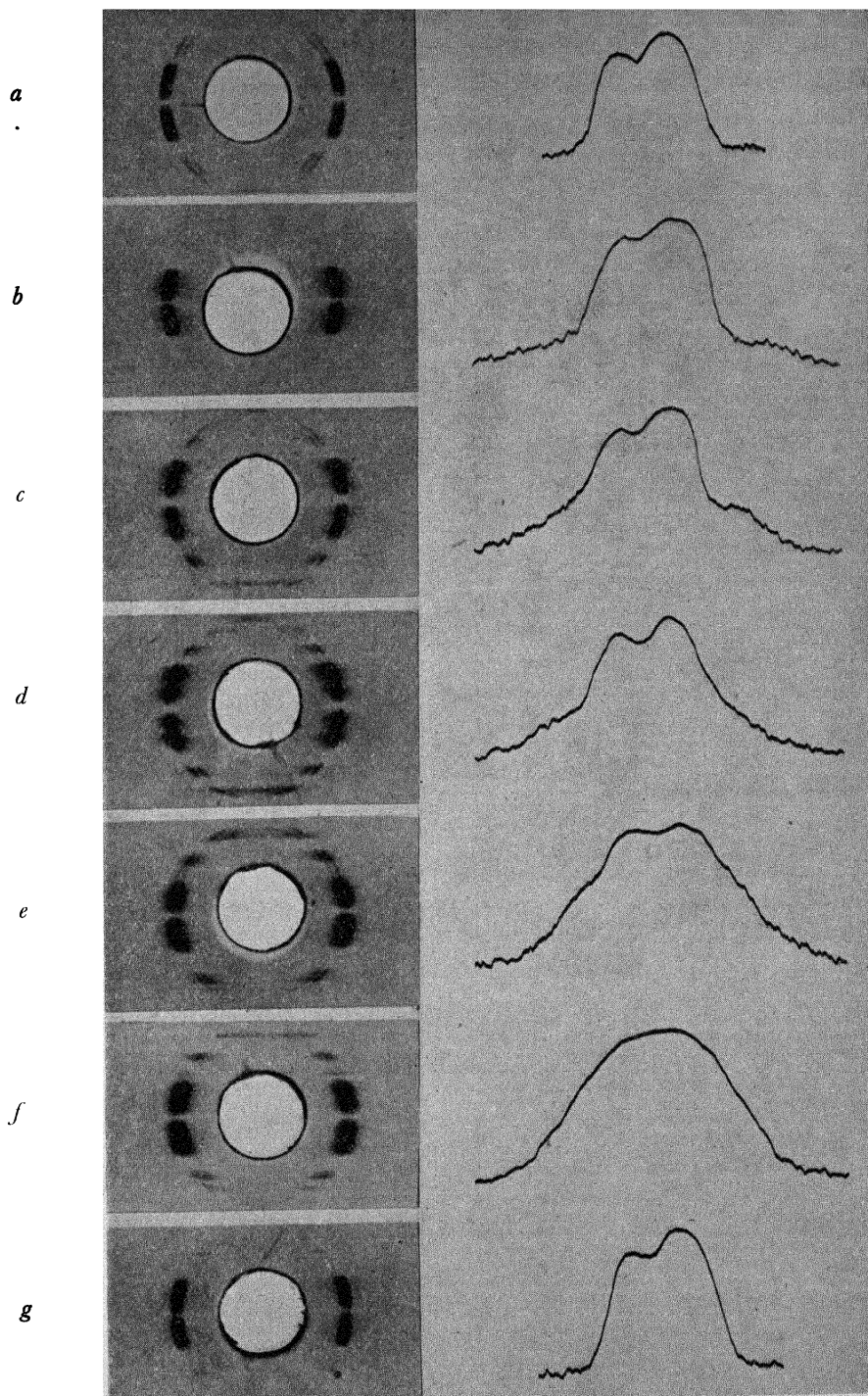
Fig. 5. Line-broadening (310 reflection; Co-Kα-doublet) in connexion with coercive force in nickel-iron-aluminium crystal. The states *a–g* correspond to those indicated in Fig. 4 by arrows. *Reproduced by permission from Physica.*

original high temperature state there is one single type of lattice, which is body-centred cubic with a clear distinction between cube centres and cube corners. This is pictured in Fig. 7 a. The nickel atoms are completely sorted out from the aluminium atoms, but the iron atoms are distributed at random, some replacing nickel, others
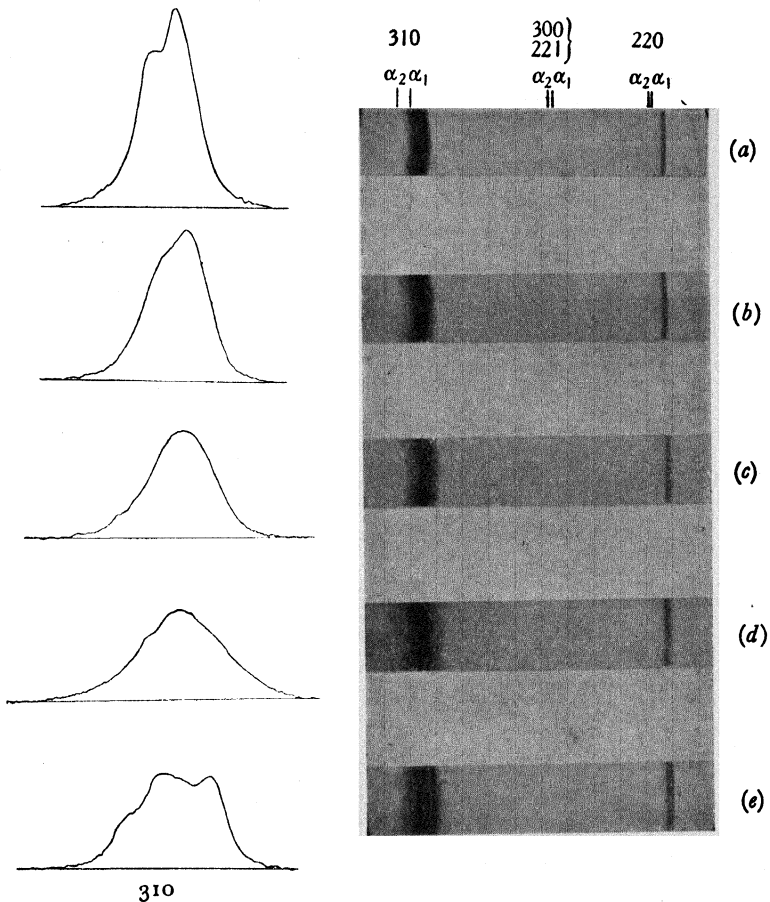

Fig. 6. Illustrating the break up of Fe_2NiAl into two phases: (a) After quenching from 900° c.; (b) after quenching from 800° c.; (c) after quenching from 700° c.; (d) after quenching from 600° c.; (e) after slow cooling.

replacing aluminium. In Fig. 7 b we show the state of the alloy after slow cooling. The crystal has split up into two portions. The smaller contains almost pure iron; the larger is very like the original lattice, but it contains fewer iron atoms.

In Fig. 7 c we show an intermediate state in which the coherence of the original lattice has not been lost, but the atomic distribution is

no longer uniform throughout the crystal. The iron atoms have begun to separate from the rest of the atoms, but at first they only form small aggregates. There is no definite crystal boundary between them and the parent crystal, and the lattice spacing is the same as in
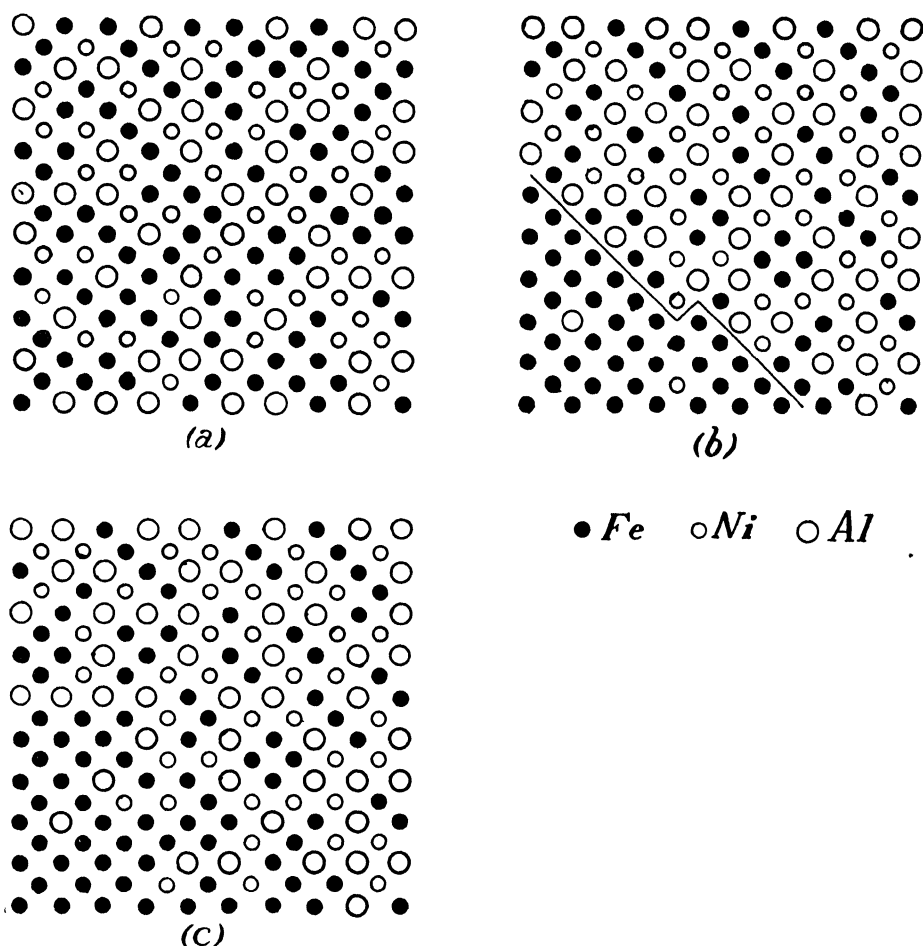

$\bullet Fe \quad \circ Ni \quad \circ Al$

Fig. 7. Fe_2NiAl after different heat-treatments. (a) Quenched 1200°, one crystal; Fe atoms at random. (b) Slow cooled, two crystals; Fe-rich crystal contracts in volume by 1 per cent. (c) Special heat-treatment for high coercivity, Fe atoms in "island"; no sharp boundary.

the original state. The small "islands" of iron cannot break away to form separate crystals once the alloy has cooled down to room temperature.

In the permanent magnetic state the alloy has the structure represented in Fig. 7 c. This state is obtained by cooling the alloy at

such a rate that the "islands" have not time to grow before the material has cooled down.

Being still forced to conform to the dimensions of the parent lattice, the iron atoms are held apart under a condition of immense strain. This is the cause of the remarkable magnetic properties of Fe_2NiAl.

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